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PROCEEDINGS OF THE CONFERENCE «FUEL CYCLES OF HIGH TEMPERATURE GAS-COOLED REACTORS»

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EDITED BY D. TYTGAT

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EDITOR'S NOTE

The organization of the Symposium « Fuel cycles of high-temperature gas-cooled reactors » was based on the assumption that all the papers were received and read by the people attending the conference a few weeks before it. Except for one paper, this was the case; the most sincere and warm thanks have therefore to be extended to all organizations which were able to assemble such extensive material in such a short time and mail it to us.

A rapporteur introduced each session, summarizing and criticizing the information presented by the various authors.

An extensive and detailed discussion was then held, interpreters having to keep pace with impulsive speakers.

A considerable amount of secretarial work in connection with the organization of this Symposium was done by Miss R. Hartwig and M. de Patoul, helped by various other members of the Euratom staff.

The first review of the taped discussion was done with Mr. De Bacci's help.

The Euratom Executive Secretariat was responsible for the logistics of the conference and thanks have to be extended to Mr. de Creeft, Mr. Breuer and their staff.

These Proceedings were edited and published by the Center for Information and Documentation (C. I. D.) of Euratom. We are very much indebted to Mrs. Stalpaert and her staff for the large amount of work done with great competence and care.

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PRÉFACE

L'Euratom participe à des recherches sur la filière des réacteurs à haute température refroidis au gaz dans le cadre de sa participation au Projet Dragon, à Winfrith, Royaume-Uni, ainsi que dans le cadre du contrat d'association avec le Projet THTR (réacteur au thorium à haute température à Juliers, Allemagne).

La conférence organisée par l'Euratom les 10 et 11 juin 1965 avait pour but de permettre à des représentants d'organisations publiques et privées, appartenant ou non à la Communauté, d'établir le bilan du développement des éléments combustibles destinés aux réacteurs en question. Les documents présentés ont porté
— sur les méthodes et les coûts de fabrication, de retraitement et de refabrication;
— sur l'incidence économique des taux de combustion et de conversion;
— et sur l'utilisation du plutonium comme combustible.

Les discussions ont mis en évidence deux points essentiels :
— sur le plan économique il est justifié de poursuivre le développement des réacteurs à haute température refroidis par gaz comprimé;
— les progrès accomplis dans la réalisation des deux projets Dragon et THTR permettent de penser qu'il faudra sans doute très peu d'années pour passer à l'utilisation industrielle de tels réacteurs.

J. GUÉRON.

" FUEL CYCLES OF HIGH TEMPERATURE GAS-COOLED REACTORS "

P. CAPRIOGLIO and al.

Summary prepared for the September 1965 issue of Nuclear News (American Nuclear Society, USA)

The Conference discussed a wide range of papers dealing with various aspects of fuel element development, the main contributors being Dragon, General Atomic, Oak Ridge and THTR.

Session I treated "*Fabrication Methods and Costs of Fuel Elements*" : While Dragon and General Atomic's Peach Bottom have been using purged fuel elements, recent trends indicate that the unpurged fuel elements can be adopted due to the development of fission products retaining coated particles.

The first unit to be produced and which alone permits comparison between the different fabrication methods, is the *coated particle.* Two main fabrication routes are possible for the kernels :

1) the powder-metallurgy processes,
2) the sol-gel processes.

1) *The powder-metallurgy processes* : Basically these processes use a mixture of Th and/or U fine oxide powders with carbon powders and a suitable binder, and have been the most used up to now by different laboratories and industries; while they use relatively simple and inexpensive apparatus, they however proceed slowly (which is a disadvantage when considering the production of large quantities of fuel), and the large amount of handling and observation might especially be annoying and costly in a remote operation.

Dragon, anyhow, is confident on the possibility of remotizing the powder metallurgy process for the contact fabrication (the process bottleneck is the granulation step).

2) *The sol-gel processes* : They are particularly appropriate for remote fabrication of thorium fuel containing U-232 and might be suitable to produce Pu fuels, but outside Oak Ridge there is no extensive production experience in the use of this process; some of the disadvantages, like handling of large quantities of waste solutions, may balance the distinct advantages that should make those processes the preferred ones in the long run, while direct comparison of the dry and wet methods is not possible yet to-day.

One question which is still open is to what extent sol-gel might be more economical than dry methods for a primary fabrication. The kernels might be made of carbides or oxides. *Carbides* have been well proved up to now. For the *oxides* the

concern is their basic thermodynamic instability in presence of carbon in the fabrication and operational conditions, but :

— low permeability coatings are applied satisfactorily as indicated by ORNL, GA and UCC,

— when the coatings fail, the fission gases release is not higher than with broken carbide particles,

— the CO release is negligible for the primary circuit, because if breakage occured, it would be progressive.

Irradiation of PyC coated UO_2 is underway at ORNL and at 18 a/o burn-ups, a fission gas release of 2×10^{-7} for Kr-88 at 1400° C has been obtained. ORNL thinks that the use of oxides should introduce savings between 5-15 % on the total fabrication cost due to a greater flexibility in the production by operation (no hydrolysis characteristics as for the carbides, no need of an inert atmosphere), but Dragon reckons the cost savings much smaller (about 0.1 %).

The coatings are mainly applied in fluidized beds (about 5'' diameter), which scale up easily especially if thorium particles can be handled separately from the uranium containing particles (which is a trend encouraged by the physics calculation). Nukem has indicated recent favorable results obtained by coating under reduced pressures.

No conclusion was obtained on the use of a *SiC coating* layer mainly favored by the Dragon Project because it leads to low uranium diffusion during coating and afterwards, and to an enhancement of the mechanical and corrosion properties of the coated particles. But long irradiation fast neutron doses data are lacking. The decision should be based on the maximum temperature and time of fuel operation and on the technique adopted for reprocessing (breaking of the particles before leaching is necessary with SiC coated particles).

On the *pyrolytic carbon coatings*, no standard specifications on coating structure, density and rate of deposition are available up to now, but work is underway in Europe and in the USA putting the emphasis on the measurement of the coatings properties to enable comparison of the production characteristics and the irradiation stability.

Recent work in Oak Ridge might open the way to very fast deposition rates (around 350 microns/hour) obtained by using large methane concentrations.

UCC and Nukem have reported their fabrication experience on *fuel spheres*. Different distributions of the coated particles are possible, even using free coated particles. Either graphitic machined shell or synthetic baked fuel elements are proposed, but the second type still needs extensive corrosion and irradiation testing.

On the use of free coated particles, no general agreement was obtained, if the interest of this concept for reprocessing is certain, but *coated particles* release in the primary circuit might prove annoying. The easiness in fabrication is still to be proved, as different geometries compared to existing concepts should be used. Further thermal conductivity measurement should indicate the temperature increase due to the absence of a graphite matrix.

Fabrication costs : Limited information has been given. General Atomic thinks that fuel, for 1,000 MWe capacity reactors, can be fabricated in the next few years for about 200 dollars/kg of heavy metal for the particles, plus about 500 dollars per element for graphite and assembly. The only detailed financial evaluation has been made by Dragon in an extrapolation of its experience gained during the manufacture of fuel for the first charge of the Dragon Experiment. A fuel fabrication capacity has been chosen with a design output of 1135 kg U-235/year. (Th/U = 10) capable of fuelling a 2,000 MWe program.

Based on specific assumptions, the estimated fabrication cost for coated particles is (excluding enriched uranium and any profit) $ 120/kg heavy metal. The total fabrication cost of a prismatic fuel element is $ 235/kg heavy metal.

Session II treated "*Reprocessing and Refabrication Methods and Costs*" : Taken as a whole, reprocessing and refabrication of HTGR fuels is an entirely new problem for which existing experience on other power reactor fuels offers limited help.

There are three successive steps :

a) Head End : removal of the uranium and thorium from the bulk graphite,
b) Purification : decontamination of U and Th from fission products,
c) Refabrication.

Several new factors must be considered :

1) The large amount of graphite per unit weight of heavy metals (U + Th),
2) The presence of the U^{232} decay chain causing a rapid growth of gamma activity in both U + Th products,
3) The extremely high specific activity met during the decontamination step.
4) Quick refabrication is necessary to minimize the growth of gamma activity in the product if high decontamination process had been adopted.

Integrated versus centralized plant : The majority of opinion supported the centralized reprocessing and refabrication plant concept. A low capacity integrated plant serving single reactor unit on the reactor site could however be conceived for special fuel elements; this could be the case of those elements in which the particles are easily separable from the graphite sleeves.

Head-end processes — Burn-leach versus grind-leach head-end processes : Silicon carbide coated particles are unaffected by combustion methods. Grinding, followed by nitric acid leaching is conceptually the simplest way in this case to bring in solution both fissile and fertile material : however, the process has not been demonstrated on actuel elements. Burning seems to be generally preferred : in this case, however, rather elaborate off-gas clean-up systems must be devised.

Solvent extraction versus fluoride volatility : The well demonstrated, thorex based, solvent extraction technology heavily unbalances the choice in the direction of aqueous methods. Furthermore, non aqueous methods inevitably bring to discard thorium with the fission products.

Powder metallurgy methods versus sol-gel techniques in the refabrication step : Dragon experts are confident that the remotization of the well established powder

metallurgical approach for particle preparation is possible and feasible with minor difficulties. This opinion has been challenged by sol-gel partisans. It was, however, agreed on the fact that sol-gel processes have a better potential and are more easily amenable to industrial operation.

Remote and semiremote versus direct fabrication : A general consensus of opinion on the necessity of semiremote or remote fabrication of recycled uranium, was reached.

Economic evaluation : The economic evaluation is very difficult because the translation of capital cost figures into unit costs depends critically on the plant size. With a plant daily through-put of approximately 4 tons of graphite, 400 kg Th + U, 36 kg of fission products and assuming the same financing as for the Nuclear Fuel Services plant, Oak Ridge has estimated an expense of 75-85 $/kg heavy metal for head-end and reprocessing.

The refabrication cost analyses made in ORNL indicate costs varying between 300 and 100 $/kg heavy metal corresponding to a plant capacity between 60 and 1,000 kg heavy metal per day. ORNL is building the Thorium-Uranium Recycle Facilities (TURF) to demonstrate the refabrication technology.

In conclusion, first results for reprocessing and refabrication are encouraging because the cost penalties of recycling fuel do not look to be unbearable. But much development and assessment work is still required.

Session III treated "*Range of Burn-Ups and Conversion Factors — Economic Assessment*" : The fuel cycle work covered once-through cycles and cycles with reprocessing. Arrangements where fuel and fertile materials were intimately mixed and seed and blanket type arrangements were discussed. The problem of excessive neutron losses in U-236 was avoided by a fuel management whereby segregation of Th and U in different coated particles was used.

There was general agreement on neutron physics results and optimum approximate operation conditions with thermal power densities between 5 and 10 Watts/cm^3 and values for specific power of the order of 2,500 KW/kg of fissile material. The estimates on fuel fabrication cost varied considerably and most authors studied also the influence of this quantity on fuel cycle cost. Because of different assumptions the results of the different authors are sometimes difficult to compare.

The principal reason for the interest in the Th fuel cycle is the high effective eta value of the uranium-233 which is relatively insensitive to neutron spectrum changes, compared with U-235 or Pu-239. Thus a good neutron economy can be guaranteed even with the hard spectrum in High Temperature Reactors which are graphite or BeO moderated. Thus the concept under consideration can be characterised by the following properties :

a) high conversion ratio,

b) high thermodynamic efficiency,

c) high specific power.

These three good characteristics cannot be achieved independently one from another and an economic optimization has to be made according to the actual cost of fissile and the actual cost of reprocessing and refabrication.

Different approaches to this problem of optimization are possible.

a) Converter on a once-through system : below a certain electrical capacity (quoted by Dragon as 2,000 MWe), there is no justification for building a reprocessing and refabrication plant. The first generation of HTGR might have to work on a once-through cycle, whereby spent fuel is discharged without credit.

This gives the opportunity of optimizing the fuel cycle for the highest possible burn-ups metallurgically achievable ($\pm$ 150,000 MWd/t), with considerable advantage to the fabrication cost, thus minimized. The disadvantages of this sort of cycle are that the accumulation of fission products in the core and disposal of spent fuel still containing some fissile are responsible for a bad neutron economy. The conversion factors achievable ($\pm$ 0.7) are therefore far beneath the potentialities of the HTGR.

The expected fuel cycle cost is :

— for Dragon 1.2-1.3 mills/kWh for a two-zone core (feed and breed),

— for ORNL 1.0 mills/kWh with spent fuel discarded and 0.885 mills/kWh in the case U-233 is credited.

b) Converter with reprocessing : this approach is justified if one assumes breakthrough of HTGR on an industrial scale, which introduces the need for reprocessing and refabrication plants. Fuel cycle costs vary between 0.88 and 1.2 mills/kWh depending on the different economic assumptions and the total capacity installed. Conversion ratios between 0.85 and 0.90 are obtainable with burn-ups between 50-60,000 MWd/t.

c) Breeding cycle : in order to be able to exploit the HTGR potentiality of breeding, some improvement must be made on the neutron economy and different core structures must be considered :

1) adoption of BeO spines within the fuel element,
2) adoption of a fission product releasing fuel,
3) elimination of control rods,
4) reduction of neutron leakage by adoption of a Th blanket, surrounding the reflector or replacing it,
5) depoisoning of the solid fission products : a reprocessing and refabrication plant designed for low hold-up time (only 10 % of the fuel outside the reactor) could be integrated with the reactor plant.

The highest potentiality towards breeding seems to belong to the pebble-bed concept, which has an unmatched flexibility of the fuel management. For a 1,000 MWe system, a conversion factor of 1.1 is attainable, even keeping the average fuel element burn-up as high as 30,000 MWD/t heavy metal. With the quoted rating of 1 MWe/kg fissile, this corresponds to a doubling time of around 15 years.

Part of the Session was dedicated to the problem of mined fissile requirements of HTGR in connection with the best utilization of natural resources. The HTGR is very well suited to satisfy future power demands, considering the limited amount of cheap uranium and the power forecast of the USA, due to the high thermodynamic efficiency of the system and the reasonably long residence time of the fuel in the reactor compared with the reprocessing time. In comparison with the Light and Heavy Water Reactors, the HTGR has the lowest fuel requirements up to 2,020. The influence of ore cost on fuel cycle cost is studied for the three converters LWR, HWR and HTGR. Here again the HTGR compares favorably with the two others. Taking 1.7 mills/kWh as a critical fuel cost — to be competitive with fossile fuel power stations — critical ore cost for LWR would be at about 10 $/lb, for HWR at about 15 $/lb, and for HTGR higher than 40 $/lb. The HTGR is in the position to use the large uranium and thorium resources available at recovery cost between 30 and 50 $/lb. The estimated cost for recovery of uranium from sea water is well in the range for economical use in an HTGR. Changing ore cost from 10 $/lb to 40 $/lb would lead to an increase from 1 mill/kWh to about 1.6 mill/kWh.

In a quick expanding industry, the adoption on a large scale of fast breeders reactors instead of advanced converters would not prevent running out of cheap uranium resources, as they would be more demanding on mined fissile, due to their poor ratings; high ratings are, in this case, more important than high conversion factors.

Session IV treated the "*Use of Plutonium as Initial Fissile Investment and/or as Make-Up Fuel*" : The interest in the use of Pu for fueling thermal reactors comes from the recognition that Pu production is increasing in the civilian reactor programs of several countries, providing a source of fuel that must be utilized. There are two basic ways to use Pu in an HTGR, namely :

1) with a complete core replacement scheme,
2) as make-up in a "feed and breed" scheme.

1) *Complete core replacement scheme* : The high termal cross section of fissionable Pu isotopes — about 3 times higher than the U-235 one — tends in principle to give better ratings to Pu fuelled reactors than to U-235 fuelled reactors. This is, however, subject to the condition that the reactivity lifetime curve has to be reasonably flat in order that reasonably long lifetime can be achieved without having to provide huge reactivity excess at the beginning of the life. To this effect, the presence of Pu-240 in substantial concentrations (15-30 %) in civil plutonium extracted from spent fuel of nuclear power station, is a most helpful feature. Pu-240 acts in fact at the same time as a burnable poison and as fertile material. By choosing appropriate particle size (Pu-240 self shielding) and by zoning the core, not only extremely long reactivity lifetime can be achieved for a relatively small initial reactivity excess, but power peaking in the core can be kept to a minimum.

Due to the high thermal capture cross section of Pu-240 which is converted to Pu-241, the total amount of fissile in the core does notc hange appreciably during

the lifetime until all Pu-240 has disappeared. After that, the reactivity drops suddenly. From this, two facts appear evident :

a) U-238 as fertile material is more suitable for this particular cycle than thorium. Thorium would in fact give place to U-233, whose thermal fission cross section for the HTGR neutron spectrum is about 3 times lower than Pu-239, and would not therefore increase appreciably the reactivity lifetime due to conversion of the additional fertile material.

b) Very little Pu will be present in the spent fuel and its reprocessing would not be worthwhile. U-233 could be instead present in appreciable quantities in case Th was used as additional fertile material. In this latter case, it might be worthwhile to reprocess the spent fuel in order to recuperate the U-233, which could be recycled or sold.

Reactivity lifetime achievable are of the order of 600 full power days, with an initial K_{eff} of the order of 1.075. Fifa values of the order of 1.3 are typical.

The fuel cycle costs would be lower than those from U/Th fuels at any fissile Pu cost below the cost of 93 % enriched U-235. As an example, Dragon has calculated that the fuel cycle cost for a U/Th once-through cycle with yearly replacement of 1/4 of the core, assuming a U-235 cost of 12 $/g would be of the order of 1.6 mills/kWh.

The same cost would be achieved by a Pu/U-238 fuelled reactor, assuming a Pu cost of 12 $/g fissile, but with a management scheme whereby the complete core would be replaced every 2 years. This fuel management scheme, which is possible only on a Pu fuelled reactor, is indeed extremely attractive and could entail substantial capital savings on charge-discharge equipment.

2) *Pu make-up in a "feed and breed" scheme* : The "breed" elements would still work on the U-235/Th-232/U-233 cycle, but the "feed" elements would contain Pu instead of U-235. At equal heat generation rate in the "feed" fuel elements would correspond a fissile Pu amount of about 1/3 of the corresponding amount of U-235. This would also be satisfactory from a reactivity point of view, considering that the eta values for U-235 and Pu are not very different.

The adoption of Pu would bring a gain of about 3 in the rating of the "feed" part of the core. This would mean a non negligible saving in the fuel inventory charges and higher costs of fissile Pu than U-235 could therefore be afforded in order to obtain the same kWh cost. This cycle has not yet been thoroughly investigated and deserves very close attention.

Some technical problems were discussed :

a) The accuracy to which predictions of reactor performance can be made : experiments in Zenith (Winfrith, U. K.) have shown that, in the range Pu/C ratios 2,420 to 14,500, nuclear data and computational techniques are adequate for practical design purposes. Attention was drawn to the fact that these experiments were carried out with a low plutonium-240 content (2 ½ %) and plans have been made to perform similar experiments with a higher plutonium-240 (26 %).

b) Moderator temperature cœfficients of reactivity were discussed and it was felt that a satisfactory negative temperature cœfficient of reactivity could be achieved. There was some doubt expressed as to how this cœfficient may vary throughout life, but the calculations carried out to date provided indications that there is no major technical feasibility point associated with this.

c) On the materials side, the importance of fuel heterogeneity was discussed when it was stated that the effect on the reactor physics of the fuel form would be far more pronounced than in a system fuelled with uranium-235. The range of fuels now being discussed was now very wide and hope was expressed for some kind of standardisation. It was felt, however, that assessment had not progressed sufficiently to allow a reduction in the possible fuels to be made.

Conclusion :

As far as HTGR are concerned, Pu stands very well in competition with a relatively expensive fissile like fully enriched U-235. Prices as high as 12 $/g fissile, or indeed even more, should the cost of fully enriched U-235 climb up in future, could be afforded for Pu. This could make of the HTGR the best bidder for the Pu stocks that are being produced.

A conclusion to be drawn from all the discussions is that it was clearly demonstrated that the fuel cycle costs of HTGR reactors are promising enough to justify further development into the industrial applications. The possibility of using plutonium in a sound economic way was particularly stressed as a very appealing feature of this reactor concept.

THE COST OF COATED PARTICLE FUEL FOR A HIGH TEMPERATURE REACTOR PROGRAMME(*)

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ABSTRACT

In order to indicate the areas where further experimental effort is likely to prove most effective in reducing fuel costs, a cost estimate based on the experience gained during the manufacture of fuel for the first charge of the Dragon Reactor Experiment has been carried out. A typical fuel specification has been selected for detailed cost analysis. It involves the fabrication of green particles by a powder metallurgy route, reaction sintering and coating with a complex layer of pyrocarbon/silicon carbide/pyrocarbon deposited in a fluidised bed. If fuelled compacts are to be employed, the coated particles are overcoated with an appropriate matrix material and pressed directly to the required shape. The detailed cost estimate is based on feed of such a fuel in a 2,000 MW(e) reactor programme.

1. — INTRODUCTION

The OECD High Temperature Reactor Project (Dragon) is a co-operative venture between twelve European countries [1]. As originally conceived the Dragon Project had two main objectives, one to build a 20 MW experimental High Temperature Gas-Cooled Reactor, the other to carry out the necessary research and development work in the field of high temperature gas cooled reactors so as to cover both general problems and those specific to the achievement of such a reactor.

As part of this programme a large effort was directed towards the development of satisfactory fuel and fuel elements. In the original concept complete release of fission product poisons was desired but experiment and experience showed that this was impracticable. This coupled with the success of the parallel development of fission product retaining fuel resulted in a radical change in the fuel concept. The fission product retaining fuel and fuel elements consist of a series of barriers and sinks designed to prevent or delay the escape of fission products into the primary circuit [2]. The most important barrier is achieved on a microscopic scale by coating individual fuel particles (500 μ diameter). Such a coating must be of low permeability to gaseous fission products and must also minimise the release of solid fission products. Pyrolytic carbon, which has a permeability coefficient (K) of about 10^{-12} cm^2/s is an obvious choice as a diffusion barrier; however it is relatively per-

(*) Dragon Project Report 340, May 1965.

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meable to certain solid fission products such as caesium and strontium. One method to improve the overall retention is to place an interlayer of silicon carbide between layers of pyrolytic carbon. The silicon carbide provides a main diffusion barrier whilst the pyrolytic carbon is employed as a supplementary diffusion barrier and as a pressure vessel.

The fuel and fuel element development programme was commenced on the laboratory scale and supported by a variety of irradiation experiments which culminated in an extended loop experiment on a prototype directly cooled power reactor fuel in the Pluto Reactor at AERE, Harwell. The fractional release rate/birth rate of noble gas fission products during operation at 1350-1400° C for 173 days was between 10^{-5} and 10^{-6}. The early promise of the laboratory work led rapidly to the parallel technological development of production processes and resulted in the manufacture by the Dragon Project of the first charge of fuel and fuel elements for the Dragon Reactor. The experience gained within the Project in the development and fabrication of coated particle fuel enables further scaling up to be envisaged for the fabrication of fuel in the quantities required for a power reactor programme.

All the production work to date by the Project has concentrated on the U^{235}/Th once-through cycle and this is the basis of the cost estimate given in this paper. Alternative fuel cycles are under study and work in this field is largely covered by other papers to this Symposium [3, 4, 5]. Some comments on these other fuel cycles are given in Section 4.

It should be noted that this paper is concerned with the factory cost and not the selling price of HTR fuel. The point is intended to be covered by the use of the word « cost » as opposed to « price ».

2. — Fabrication routes

2.1. — General.

The fabrication routes for coated particle fuel are summarised in Fig. 1. They may be classified as follows :

(i) Sol-Gel processes.
(ii) Oxide routes.
(iii) Massive carbide routes.
(iv) Powder metallurgy processes.

The end product of these processes is a fuel kernel which is then coated.

The various processes are discussed in subsequent sections of this paper.

2.2. — Sol-Gel Fabrication Routes.

The Sol-Gel fabrication routes are useful in that they lead to particles which are very spherical in shape but more important they are particularly appropriate for the remote refabrication of fuel involving the thorium/U^{235} fuel cycle.

The basic procedures to make Sol-Gel particles are described in a companion paper to this Symposium [5]. In the field of oxide fuel it should be noted that Sol-Gel

oxide particles have high strength. However there is no evidence to suggest that this is important for fuel which is expected to have a high burn-up.

Present experience suggests that the particle size chosen for a high temperature reactor fuel might affect the choice between Sol-Gel and other routes, for the powder metallurgy route is more easily applied to particles in the region 500-2,000 microns diameter whilst the Sol-Gel route seems at the present confined to particles below 600 microns diameter.

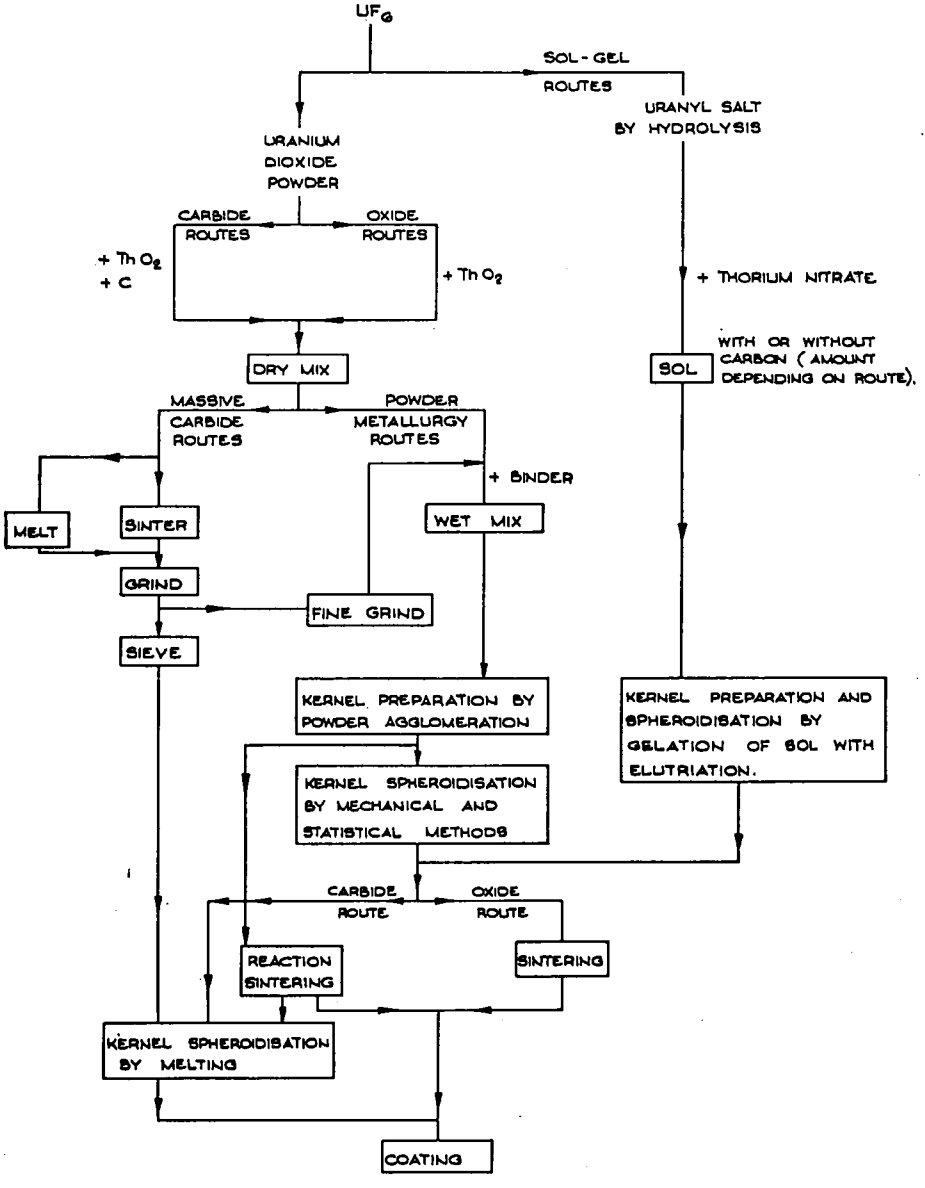

FIG. 1. — Fabrication Routes for Coated Particle Fuel (once-through Cycle).

Due to the fact that work in the Dragon Project with enriched uranium Sol-Gel particles has not yet proceeded beyond the laboratory scale and because it is intended that this cost estimate should have as realistic a basis as possible the Sol-Gel routes will not be considered here in further detail.

2.3. — Oxide Routes.

Initially the Dragon Project rejected the use of oxide particles coated with pyrolytic carbon due to fear of the effect of the reaction between uranium or thorium oxides with the carbon of the coating under operating conditions in the reactor. It appears likely that due to the extremely low permeability of the pyrocarbon coating ($K \simeq 10^{-12}$ cm^2/s) the forward reaction to carbon monoxide will be suppressed at a rather low partial pressure of carbon monoxide [6]. Thus the pressure build-up within such a particle will not be significantly increased by the reaction to form carbon monoxide. The main technical questions with such particles are :

(i) The effects subsequent to particle breakage when the oxides might be expected to be reduced by the carbon of the coatings leading to evolution of carbon monoxide resulting in mass transfer and corrosion problems.
(ii) The effects of long term fast neutron irradiation.
(iii) Whether oxide fuel kernels have significantly different retention properties compared with carbide kernels.

The present irradiation programme of the Dragon Reactor Experiment incorporates some coated particle oxide fuel. However, since our experience is still confined to the laboratory scale, the various oxide routes will not be considered here in the detailed cost estimate. Nevertheless it can be stated that, in principle, the oxide route will not differ significantly from the equivalent carbide route due to the many common components of the cost.

2.4. — Massive Carbide Routes.

Although a carbide route was used for the manufacture of the driver fuel of the first charge for the Dragon Reactor Experiment, none of these routes is preferred for the following reasons.

(*a*) With uranium/thorium dicarbide fuel the problems of handling are accentuated due to reaction with atmospheric moisture.
(*b*) An extra furnace treatment is necessary. In the first furnace treatment the carbide is melted or sintered in massive form and in the second the particles are melted or sintered.
(*c*) In one of the routes the massive carbide has to be ground to give a particular sieve range of particles. Such a procedure is inefficient even if special steps are taken to remove the correct size of particles from the grinding mill as soon as they are made, because inevitably the sieving operation will lead to the recycling of part of the product.
(*d*) Several of the massive carbide routes lead to a final spheroidisation of the particles by melting, such as in a plasma torch or by induction heating in an excess

of carbon. Although the Dragon Project has included some melted particles in its irradiation programme the main emphasis has been on particles of controlled porosity made by powder metallurgy agglomeration techniques. If melted particles are used then any increase in volume due to fission has to be met by way of porosity in the coating either by building in porosity during the manufacture of the inner coating or by irradiation shrinkage of the inner coating. In general, since melting must be carried out at a higher temperature than sintering, it should be more expensive.

2.5. — Powder Metallurgy Processes.

The powder metallurgy route requires that the particles used to make the kernel should be extremely fine such that the incidence of a particular particle cannot affect the shape or composition significantly. Once the particles are agglomerated they can be spheroidised in a variety of ways of which the more important at the present time are :

(i) plasma jet melting,
(ii) melting in a « bed » of carbon powder,
(iii) spheroidisation before any heat treatment by using either a mechanical force to shape the particles or the natural tendency of the powders to agglomerate into spheres.

As explained previously it is preferred to build the necessary porosity into the kernel. This is achieved by bonding uranium and thorium dioxides with carbon and an organic binder and reaction sintering rather than melting the kernels. Because this then rules out the utilisation of melting to achieve the necessary spheroidisation, it is necessary to spheroidise the particles before sintering. Apart from the ease with which the necessary porosity can be controlled, the main advantage of the particular route selected for exploitation within the Dragon Project lies in the definition of the shape and size of the particle before any major transformation of the raw materials is made. Thus the cost of recycle, which is inevitable when a narrow particle size range is desired, is minimised.

2.6. — Coating.

For economic reasons it is desirable that the coating thickness shall be as uniformly thick as possible. Thus a violent agitation of the particles during the disposition process is required so as to prevent either agglomeration or preferential deposition. Amongst the possible methods, fluidisation, tumbling, moving bed and vibration, only the first two are able to achieve this criterion.

2.7. — Fabrication Route Chosen for Cost Estimate.

The fabrication route chosen for detailed cost estimation is no more than a scale-up of the actual process used for making the fuel for the First Charge of the Dragon Reactor. The operations are summarised in Fig. 2.

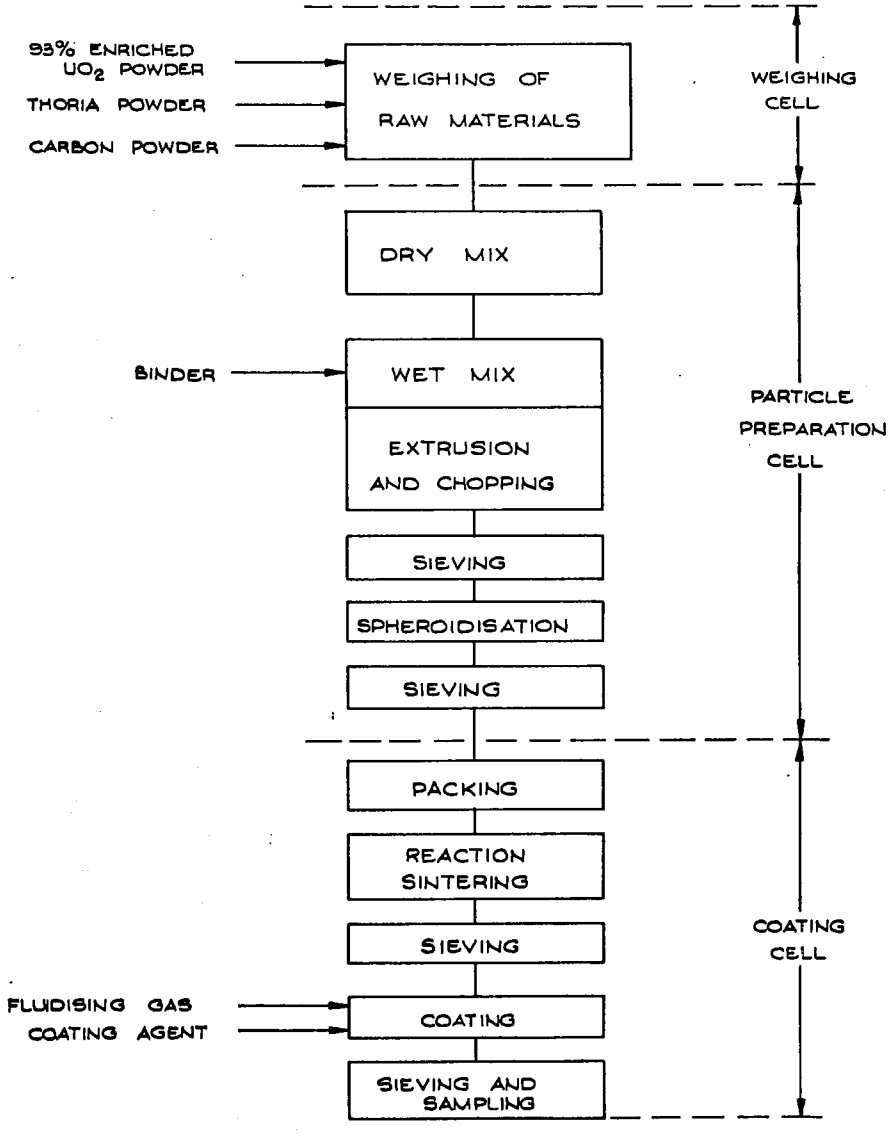

FIG. 2. — Fabrication Route chosen for Coated Particle Cost Estimate.

The raw materials are weighed in the correct proportions in a separate cell and then passed through a wall store to the particle preparation cell.

All particle preparation is carried out within a single glove box. Dry powders are mixed by a screw mixer in a conical hopper and then metered into an extruder where the binder is injected. The wet mix is extruded through fine holes as spaghetti and the approximate particle volume achieved by chopping the extrusions. The crumbs so formed are sieved and then spheroidised in a planetary mill modified

for semi-continuous operation with a minimum amount of handling. The spherical particles are sieved and transferred through a wall store to the reaction sintering cell.

The particles are packed for sintering in graphite crucibles and then passed through a semi-continuous vacuum furnace. After sintering the particles are unpacked, sieved and passed forward through a wall store to the coating cell.

In the fluidised bed furnaces pyrolytic carbon is deposited from methane and silicon carbide from methyltrichlorosilane. The fluidising gas is initially argon, then hydrogen for the silicon carbide deposition and finally nitrogen for the outer pyrolytic carbon layer.

Coated particles are subsequently sieved to remove pyrolytic carbon flakes or powder and then a sample is taken for quality control purposes.

3. — Cost Estimate

3.1. — Reference Cost Estimate.

In the previous section a simple once-through fuel fabrication route for uranium-thorium dicarbide coated particle fuel was described. This process will be the basis for the detailed cost estimate though extrapolations to other types of fuel will be attempted.

3.1.1. — *Capacity of the Plant.*

It is reasonable to assume that for the first generation of high temperature gas cooled reactors a once-through fuel cycle will be used. The upper limit for an initial nuclear power programme can be considered to be set by the level at which reprocessing of fuel would become economic and this can be deduced to be in excess of 2,000 MW(e) installed capacity [7, 8]. It seems likely that the first power reactor programme based on a new concept will not be in excess of the same figure — 2,000 MW(e). The lower limit of such a programme is obviously set by the size of reactor unit being constructed.

With these considerations in mind, for the purpose of this reference cost estimate, a fuel fabrication capacity has been chosen with a design output of 1,135 kg U^{235}/year.

This output is capable of fuelling a 2,000 MW(e) (5,000 MW[Th]) programme consisting of four reactors, one station being constructed each year.

For the case which has been chosen, with a Th : U^{235} atomic ratio of 10 : 1 ($N = 10$) the initial feed for each station is assumed to be 730 kg U^{235} the annual make-up thereafter being 270 kg/station.

3.1.2. — *Technical Assumptions.*

It is assumed for the moment that the process finishes at the coated particle stage, though projections of the cost of consolidating the fuel into a fuel element will be given in Section 3.5 of this paper. Given the assumption of 5 % recoverable rejects and 1 % irrecoverable loss the chosen throughput can be translated into a flow sheet as shown in Fig. 3.

The following technical bases have also been considered :

(i) It is assumed that satisfactory arrangements can be agreed with the licensing authority as regards the criticality hazard both in relation to movement of fissile material and its storage.

(ii) It is assumed that particle preparation can be automated as described in the previous section.

(iii) The output of sintered kernels would be achieved by reaction sintering in a single batch continuous furnace.

(iv) The output of coated particles is obtained with two fluidised bed furnaces with 6 inch diameter reactors but 50 % excess capacity is installed by means of a third furnace. The latter part of the coating operation is carried out with nitrogen as fluidising gas.

(v) The encouraging experience obtained during the manufacture of the First Charge of the fuel and fuel elements for the Dragon Reactor Experimcnt when handling 93 % enriched uranium and thorium has demonstrated that handling and contamination problems are minimal. This allows a fairly simple form of building and enables the production unit to be very compact. A T- or L-shaped building is envisaged comprising :

Office block 165 m² (1,775 ft²)
(inactive) } superficial area
Active area 650 m² (7,000 ft²)

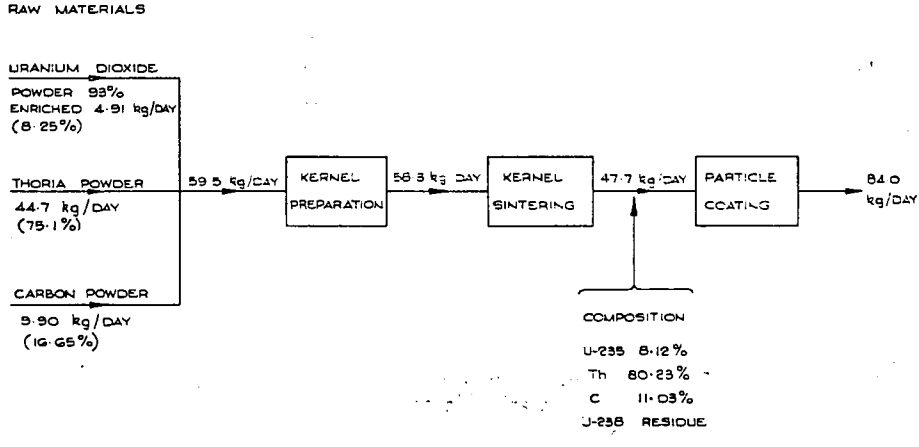

FIG. 3. — Flow Sheet for 1135 kg U^{235}/Annum Coated Particle Fuel Production Unit.

Within the active area there are four process cells (weighing, kernel preparation, kernel sintering, coating), a fissile material store, a fertile material store as well as an active workshop, a control laboratory and decontamination facilities.

3.1.3. — *Economic Assumptions and Details.*

The economic assumptions are listed below together with other economic details :

(i) The fuel production unit would be part of a larger, but not necessarily nuclear, complex whose services it employs for general items such as the personnel and labour offices, police, medical supervision, bill paying, etc. The larger complex will also provide a chemical analytical service under contract.

(ii) The production will be carried out for 300 days per annum on a 24 h basis using four shifts to run a three shift system. The labour force is detailed in Table 1.

(iii) The total hold-up in the plant is taken as 30 days, half of the U^{235} in the form of raw material with the remainder counted as coated particle fuel.

TABLE 1. — Coated Particle Production — Labour Force
(1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

Grade		No. on Shift	Total
Direct Labour — Particle Preparation	General Worker	2	8
— Sintering	General Worker	1	4
— Coating	General Worker	2	8
Supervisor	Technical	1	4
Graduate			3
Secretary			1
Materials Controller	Technical		1
Control Laboratory Staff	General Worker		1
Health Physics			1
Mechanic			1
Electrician			1
			33

Note : Annual salaries :

Graduate	£2,500
Technical Grade	£1,600
Materials Controller	£1,100
Health Physics Surveyor	£1,100
Mechanic	£1,100
Electrician	£1,100
Secretary	£800
General Worker (shift)	£900
General Worker	£800

(iv) Costs of raw materials, delivered equipment, building and services are deduced from various sources mainly within the United Kingdom.

(v) For the determination of the fixed capital investment the following corrective factors have been applied :

50 % of the delivered cost of equipment for its installation,

20 % of the building construction cost for its installation (including connections to the main complex, supplies of utilities, etc.), and

30 % of the total installed cost for engineering and design expenses.

It has also been assumed that before commencing to process enriched uranium, the production line will be worked up by a « preproduction run » whose costs will be equivalent to six months running cost without charge for the U^{235} or for the assumed U^{235} losses and rejects.

(vi) For the determination of the running costs :

The labour cost has been taken at the United Kingdom level increased by 65 % to allow for auxiliary labour charges (such as National Health Service, Pension Scheme, etc.).

Maintenance and repair costs have been assumed to be 15 % of the installed equipment costs.

The amortisation of the fixed capital has been made linearly, that relating to the building itself over 20 years with the remainder over 10 years.

The overhead and general expenses have been allocated to the different process stages according to the area occupied. These expenses include the running cost of the building and common services (heating, ventilation, maintenance, waste disposal, consumable goods, etc.), the depreciation of the building and common services, management, supervision, the labour costs of the common services, communal taxes, insurance and services from the main complex.

(vii) The financial charges have been taken to be 4 % of total capital investment (fixed + working capital).

(viii) No allowance for profit has been made nor for corporation expenses such as advertising, research and development, sales, etc.

3.1.4. — *Results.*

The capital cost of the equipment for the production unit as delivered is given in Table 2, whilst Table 3 gives a summary of the running costs for the production areas. The capital investment summary, given in Table 4, shows that the total of fixed and working capital is £1,110,000 ($3,100,000).

The total running cost, derived from Table 3, gives a total product cost per annum of £6,210,000 or, discounting the cost of the U^{235} contained in the product, an added value of £535,000 ($1,500,000). From this added value the estimated fabri-

cation cost can be expressed in terms of U^{235} content, heavy metal content (1) or as coated particles and we have :

£472/kg U^{235} . . . . . (\$1,300)
£43/kg heavy metal . . . . . (\$120)
£21/kg coated particles . . . . . (\$59)

TABLE 2. — Coated Particle Production — Delivered Equipment Cost
(1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

	£
Weighing cell	1,000
Kernel preparation cell	9,000
Sintering cell	35,000
Coating cell	30,000
Workshop	4,000
Control laboratory	8,500
Office equipment	2,500
TOTAL	90,000

3.2. — DISCUSSION OF REFERENCE COST ESTIMATE FOR COATED PARTICLE FABRICATION

3.2.1. — *General.*

Having prepared the reference cost estimate it is instructive to analyse the areas of high cost. This data is reassembled in Table 5 from which it can be seen that less than 50 % of the cost depends on the chosen fabrication route.

3.2.2. — *Coating.*

It is evident from Table 5 that the most expensive part of the process is coating. The running cost of coating can be seen from Table 3 to be made up as follows :

Raw materials . . . . . £42,750 (32.9 %)
Power and water . . . . . £13,450 (10.3 %)
Direct labour . . . . . £8,900 (6.8 %)
Maintenance, repair and depreciation on equipment . . . . . £12,600 (9.7 %)
General expenses and overheads . . . . . £52,400 (40.3 %)

On the assumption that the general expenses and overheads cannot be reduced drastically, the fruitful area for economy is in the raw materials, almost all the cost

(1) The cost is expressed in terms of heavy metal content purely for comparative purposes; it will be seen later in this paper that it is not strongly dependent on the heavy metal content.

TABLE 3. — Coated Particle Production — Breakdown of Running Costs per Year (in £ Sterling) (1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

Item	Process Stage				Total
	Weighing	Kernel Preparation	Sintering	Coating	
Materials	0	200	650	42,750	43,600
Power and Water	0	150	8,600	13,450	22,200
Direct Labour	0	14,250	6,550	8,900	29,700
Maintenance, Repair and Depreciation on Equipment	250	3,800	14,750	12,600	31,400
General Expenses and Overheads	5,900	18,200	23,900	52,400	100,400
TOTAL	6,150	36,600	54,450	130,100	117,300

Financial charges on Working Capital	25,000	see Table 4
Financial charges on Fixed Capital	19,500	see Table 4
93 % Enriched Uranium Dioxide	6,135,800	
Thoria and Carbon	67,400	
	6,475,000	
Less Recovery of UO_2 from Reprocessing of recoverable Rejects	265,000	
Total running cost	6,210,000	
Less Recovery of U^{235} in Form of Coated Particles	5,675,000	
Total added Value	535,000	

being incurred in the supply of gas and silane to the fluidised bed furnaces. The detailed gas and silane costs per annum are estimated to be :

Nitrogen	£4,370
Argon	£18,500
Hydrogen	£4,590
Methane	£3,600
Methyltrichlorosilane	£11,400

The argon cost quoted is already reduced, as compared with current practice, by substitution with nitrogen as early as possible in the coating operation. Nevertheless, because it is used on a once through basis its annual cost is high and methods of recirculation are worthy of investigation. The purification of the argon may not be difficult if hydrogen need not be eliminated and a reduction in argon cost

TABLE 4. — Coated Particle Production — Capital Investment
(1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

	£	10^3\$
Fixed Capital		
(*a*) Delivered Equipment (Table 2) £90,000		
(*b*) Installed Equipment (1.5 × *a*)	135,000	375
(*c*) Building (including Air Conditioning, Gas Stores, Active Stores, etc.) £105,000		
(*d*) Installed Building Cost (including external Piping, Wiring, etc.) (1.2 × *c*).	126,000	350
(*e*) Total installed Cost (*b* + *d*)	261,000	725
(*f*) Engineering and Design (0.3 × *e*)	78,500	217.5
(*g*) Preoperational Cost (Six Months Running with Natural Uranium instead of enriched)	145,500	404.5
Total fixed Capital Cost (*e* + *f* + *g*)	485,000	1,347
Working Capital		
10 % Annual Running Cost less Gas but including Fissile Material	617,800	1,717
Gas Supply for One Week	800	2.2
Spare Parts (5 % of installed Equipment Cost)	6,400	17.8
Total Working Capital	625,000	1,737
Total Capital Investment		
Fixed Capital	485,000	1,347
Working Capital	625,000	1,737
TOTAL	1,110,000	3,084

TABLE 5. — Coated Particle Production — Breakdown of Costs
(1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

Item	Cost			
	£/kg U^{235}	\$/kg U^{235}	% Total Fabrication Cost	
Conversion from UF_6 to UO_2	100	278	21.2	
Thoria and Carbon	59.4	165	12.6	49.2
Irrecoverable Losses (1 %)	51	141.8	10.8	
Recovery of 5 % Reject	21.5	59.8	4.6	
Coating	114.8	319	24.3	
Sintering	48.1	133.8	10.2	
Kernel Preparation	32.4	90	6.9	50.8
Weighing	5.5	15.3	1.1	
Financial Charges	39.3	109.3	8.3	
TOTAL	472	1,312	100.0	

in excess of 90 % could then be anticipated. Against this, however, one must poin out that some purification plants tend only to move the area of cost from that of raw materials to that of maintenance. Since operation with hydrogen is necessary for the satisfactory deposition of silicon carbide from methyltrichlorosilane, the additional hazard from a build-up of hydrogen in the recirculated argon may be acceptable. Extending this argument a little further, it may be considered possible to eliminate completely the use of argon and replace it by hydrogen, nitrogen or any cheaper inert gas mixture.

It is obvious that a considerable amount of money is expended in fluidising the particles and because of this an examination of alternative methods of particle agitation is suggested. For if all « fluidising gas » costs could be eliminated then the cost of coating quoted in Table 5 would be reduced by about 20 %.

Four general methods of coating have been outlined earlier but apart from fluidisation thc only other process worthy of consideration is a rotating furnace or tumbling bed.

Although the tumbling bed avoids the use of fluidising gas it has other limitations. These stemming directly from the need to keep soot formation to a minimum, are a function of the cooled gas injection system. In consequence :

(*a*) The bed depth is restricted to about one third of the cross sectional area, otherwise particles would be cooled by their proximity to the injection system.

(*b*) The power consumption is increased to the «cold finger» right through the furnace.

(*c*) The deposition rate is relatively low.

(*d*) The type of coating cannot be varied over wide limits.

Possibly the further development of the tumbling bed allowing greater flow rates would affect the comments under (*c*) and (*d*) above, but this is not yet clear.

Thus we may conclude that the tumbling bed method is likely to be a slower process with less output per unit volume of furnace hot zone. Two other possible disadvantages are that the tumbling action may generate fine particles which are not carried out of the bed and that soot problems are likely to be aggravated due to the large free volume of gas.

Of the raw material cost, that attributed to methyltrichlorosilane is easily the second most expensive. From this point of view silane is about ten times more expensive than methane per unit volume of coating. However the deposition rate is slightly higher but this will only affect the cost marginally. Whether or not the silicon carbide interlayer is advantageous is primarily a technical decision based on long term irradiation results, which therefore cannot be resolved at the present time. It should be noted however that the silicon carbide interlayer renders reprocessing of coated particle fuel slightly less flexible as it vitiates a simple oxidation route and demands a crush/leach method.

3.2.3. — *Conversion from Uranium Hexafluoride.*

After coating, the next most expensive item is the cost of conversion from UF_6 to UO_2 at £100/kg U^{235}. For the very small quantity required for the Dragon

Reactor a much higher price was charged, though, were the throughput in the particular conversion plant to be increased to about 1,200 kg U^{235}/annum the price would reduce to £150-200/kg U^{235}. A further reduction to near £100/kg U^{235} would be expected if a new plant were to be installed. This figure is in good agreement with offers of U^{235} from the United States of America when allowance is made for the cost of transport to Europe. One verbal estimate from a European firm is at the same price level. Thus there is no basis for projecting a cost much different from £100/kg U^{235}.

Yet because it is such a large proportion of the cost of making coated particles the case for studying the conversion from UF_6 in greater detail than is possible in this paper is evident. Thus the presence of residual fluoride in the oxide is probably of no consequence when it is to be reacted to carbide. As a second example there are, in principle, methods of proceeding directly from hexafluoride to carbide.

3.2.4. — *Thoria and Carbon.*

The cost of thoria and carbon is dominated by the cost of thoria, which in this study was taken to be £5/kg ThO_2. This is 25 % higher than the value used in the Advanced Converter Study [9] but over 20 % lower than that paid for the relatively small quantities employed in the Dragon Reactor. Taking the figure given in ORNL-3686 would reduce the cost under this heading by £11.8/kg U^{235} to £47.6/kg U^{235}.

3.2.5. — *Losses.*

The fourth most expensive aspect revealed by the reference cost estimate is the irrecoverable losses. These were arbitrarily assumed to be 1 % against 0.2 % taken in [9] but, with our own experience of fuel fabrication processes and, in advance of large scale production, we have no confidence in a figure lower than 1 %. We have further evidence from uranium carbide production (other than for the Dragon Project) that a value of 1 % is reasonable.

Indeed when one considers that this loss figure is intimately connected with the errors of weighing, sampling and analysis, further support for the higher figure is found.

3.2.6. — *Conclusions Regarding Reference Cost Estimate for Coated Particle Fabrication.*

The four items discussed above account for approximately 70 % of the fabrication costs. The remainder of the cost is spread over several process areas and large reductions in cost (other than by increasing the scale of operations) are not to be expected. It should also be noted that the cost of recovering the 5 % reject is based on processing of this material away from the production unit. If silicon carbide is not used as a coating interlayer, then there are indications that direct oxidation of this material back to UO_2 in an extension to the production unit would be advantageous. With silicon carbide coatings however, a crush/leach/precipitation/fire method would be involved.

In this discussion various possible modifications to the fabrication cost have been suggested. These are summarised in Table 6. Since large uncertainties are involved taking us beyond the existing technological position this table should be regarded as somewhat speculative.

TABLE 6. — Coated Particle Production — Cost Projection
(1,135 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

Item	Cost (£/kg U^{235})	Remarks on Assumptions
Conversion from UF_6 to UO_2	100	
Thoria and Carbon	47.6	Thoria cost as used for advanced converter study (ORNL-3686).
Irrecoverable Losses (1 %)	51	
Recovery of 5 % Reject	5	Either negligible loss in coating or SiC interlayer assumed to be technically acceptable.
Coating	88.5	Nitrogen as fluidising gas, elimination of SiC interlayer assumed to be technically acceptable.
Sintering	48.1	
Kernel Preparation	32.4	
Weighing	5.5	
Financial Charges	39.3	
TOTAL	£417.4 ($1,160)	

3.3. — INFLUENCE OF THROUGHPUT ON COST.

It is assumed that production is always carried out using similar equipment, the variation in throughput being achieved either by a reduction in the size of the equipment or by a reduction in the hours worked per day.

It is evident that the specific cost is made up of two terms one of which can be assumed to be independent of throughput, with a second variable term dependent upon throughput. The first term will consist of the following items from Table 5.

	£/kg U^{235}
Conversion from UF_6 to UO_2	100
Thoria and carbon	59.4
Irrecoverable losses	51
Recovery of 5 % reject	21.5
Financial charge on working capital only	22.5
Raw materials such as gases for coating	38.4
Total	£292.3

The sum of these items gives the approximate asymptote of the fabrication cost for infinite throughput using similar equipment. The calculations of the reference cost estimate have been re-run with the throughput varying from 15 to 1,135 kg U^{235}/annum and the results of this investigation are given in Fig. 4. For the case of 380 kg U^{235}/year throughput, two modes of working were assumed, single shift and 24 h shift respectively. It is interesting to note that the extra cost of single shift working is only about 7 % overall. The explanation for this is that since the equipment required for production is relatively small its price will not vary much with throughput whilst the general expenses will remain at about the same level.

For low production rates the assumption of a fixed term totalling £292.3/kg U^{235} is obviously optimistic but the error at 15 kg U^{235}/annum throughput is only about 13 %. In any case the assumption of a fixed term makes the effect of the variable terms more evident.

3.4. — Effect of Variation in Thorium/Uranium^{235} Ratio on Cost.

The effect of varying the Th : U^{235} atomic ratio (the N-value), with the U^{235} throughput maintained at 1,135 kg/annum, has been studied for three cases in addition to the reference cost estimate, i.e.,

(*a*) N — 5, homogeneous core.
(*b*) UC_2 fuel, N = O.
(*c*) ThC_2 fuel, N = ∞.

In the pure UC_2 case the batch size has been assumed to be limited in order to reduce the effect on cost of a complete batch failure. For this reason, in this particular case, the batch size has been limited for room temperature processes to 1.3 kg U^{235} and 0.650 kg for furnace operations. Then in order to estimate the cost of a feed and breed system the costs for UC_2 fuel and ThC_2 fuel have been combined.

The results of these calculations which are shown in Fig. 5 show a linear dependence with N-value at least up to N = 10. It should be noted that to simplify Fig. 5

the curve for the feed and breed case assumes that the UC_2 and the ThC_2 fuel are manufactured in separate plants. A combined plant for manufacturing both feed and breed with common overheads would, at the N = 10 level, result in a saving of about £50/kg U^{235}.

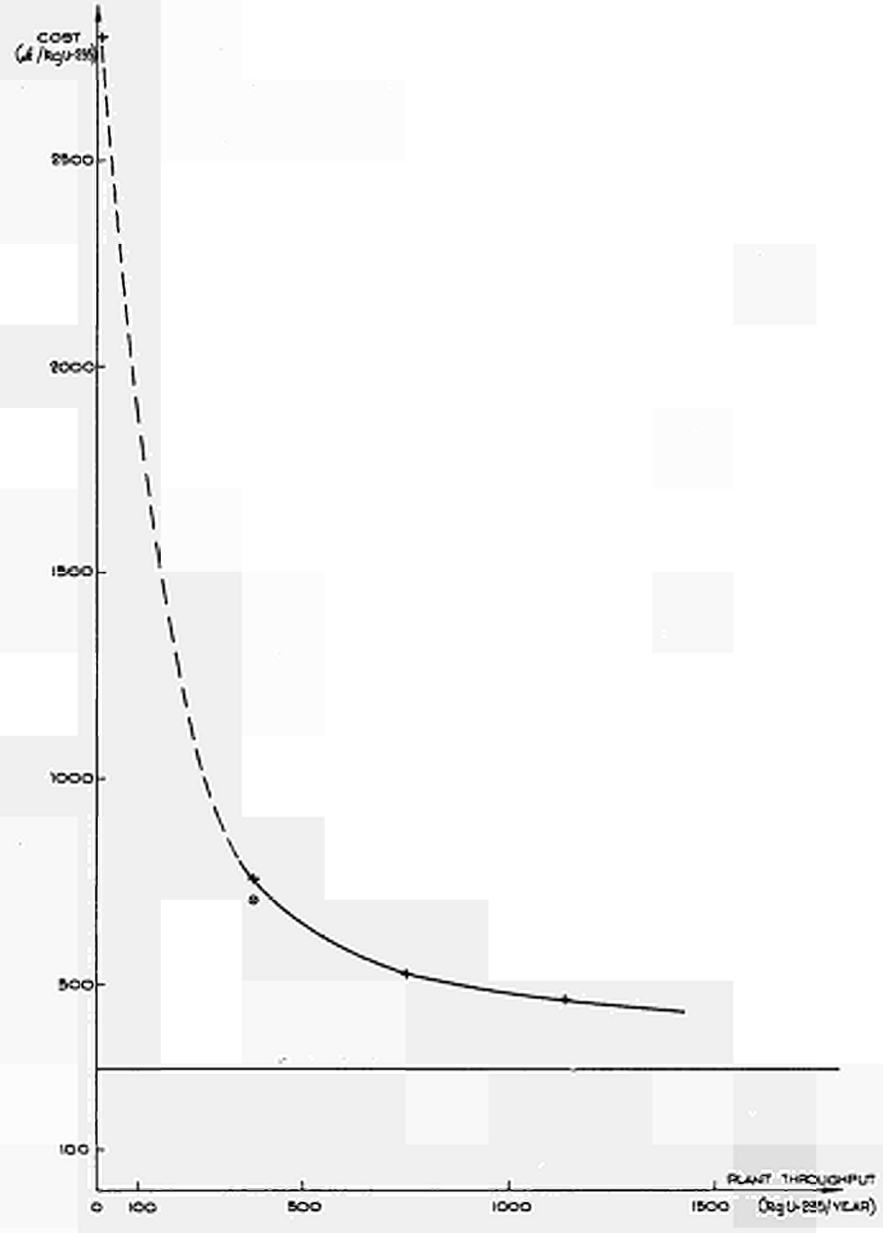

FIG. 4. — Influence of Throughput on Cost for Fixed Th/U^{235} Ratio.

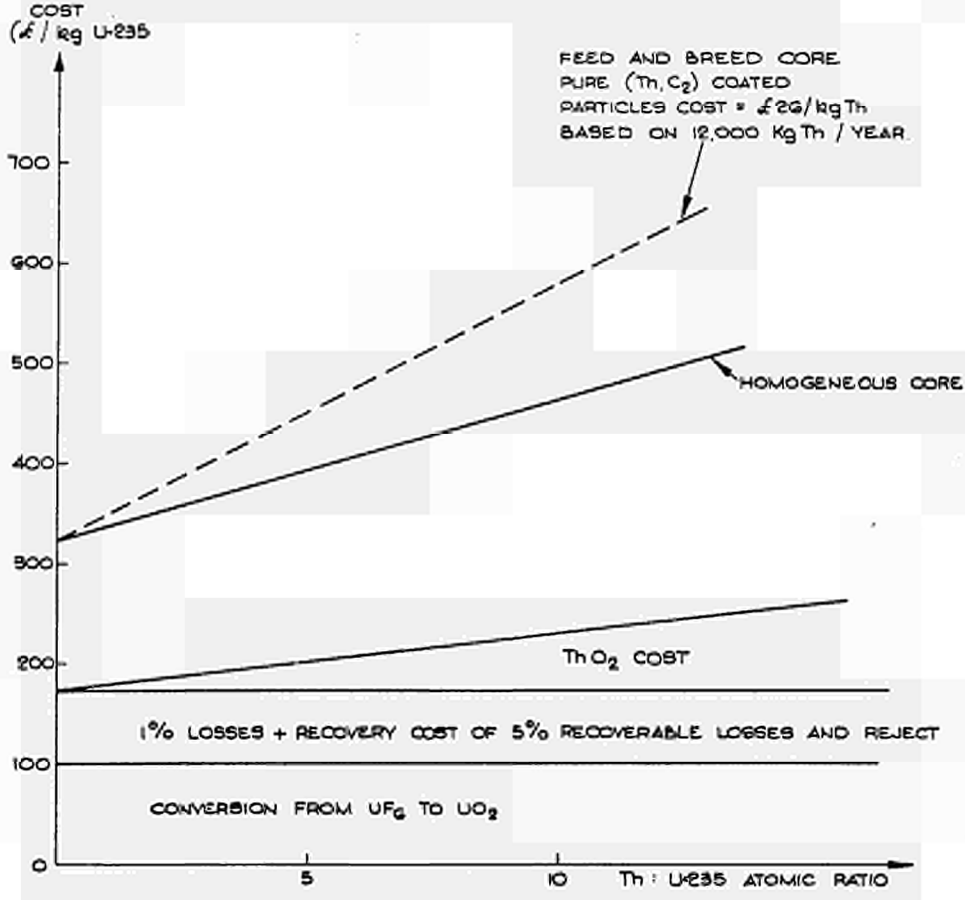

FIG. 5. — Cost of Coated Particle Fuel Versus Th/U^{235} Ratio.
(Plant Capacity fixed at 1135 kg U^{235}/Annum.)

3.5. — COST OF CONSOLIDATING COATED PARTICLES INTO FUEL ELEMENTS.

3.5.1. — *General.*

In this section the additional cost of consolidating coated particle fuel into a fuel element will be estimated. The reasons for separating the cost in this way are twofold :

— First there are several vendors of coated particle fuel.

— Secondly there are large divergences in the design of HTR fuel elements which are likely to have a significant effect on cost.

As an example of one type of design there is the « telephone dial » arrangement of the General Atomic Target Study as exhibited at the 1964 Geneva Conference. In this approach coated particle fuel is simply poured into a series of holes in the wall of a long graphite tube. Although there are difficulties caused by the aspect

ratio of such a design, the concept of using loose particles will be retained as an illustration of what should in principle be one of the cheapest methods of consolidation.

Doubt about the validity of using loose particles in fragile graphite blocks leads to the compaction of the coated particles into a matrix and then the insertion of the fuelled cylinders or spheres into graphite components. The use of some type of matrix is common to the AVR Pebble Bed Reactor, the Peach Bottom Reactor and the Dragon Reactor Experiment.

3.5.2. — *Cartridge Route.*

The consolidation of coated particles into a matrix and the assembly of the fuel element is assumed to be carried out in an extension of the coated particle production building. The additional superficial area required is estimated as follows :

430 m^2 active.

50 m^2 inactive.

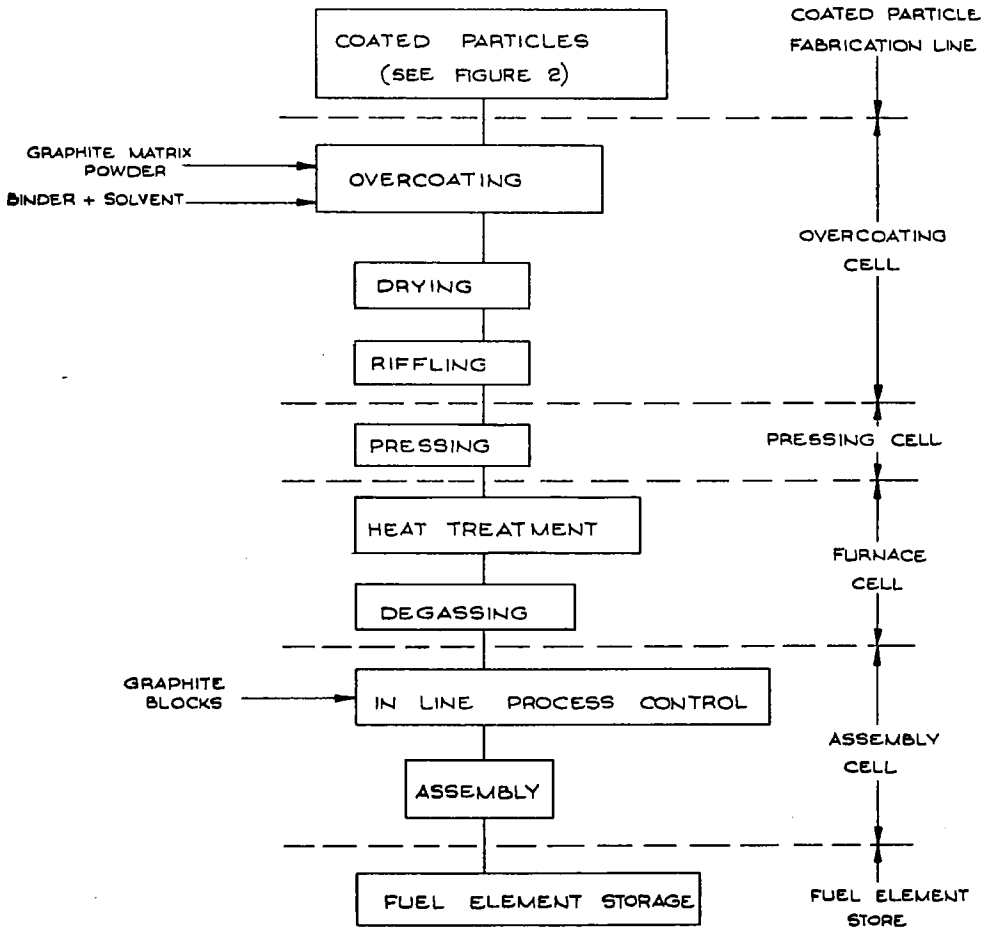

FIG. 6. — Fuel Element Manufacture.

The consolidation line would be placed parallel to the coated particle production line and using its output of 1,135 kg U^{235}/annum in the form of coated particles with the common services of active workshop, control laboratory and decontamination between the two lines.

Fig. 6 gives an outline of the consolidation process. The additional direct labour force required is five men per shift, again using four shifts to operate a three shift system for 300 days/year with two men on day work. The five shift workers are employed in the semi-continuous processes of overcoating, pressing, heat treatment and outgassing whilst the day workers carry out the in-line process control (of cartridge dimensions, gross defects and U^{235} content), assembly and fuel element storage. There is one additional supervisor in charge of each shift.

TABLE 7. — Consolidation — Cartridge Route — Capital Investment
(1,110 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10 starting from coated particles)

	£	£	10^3\$
Fixed Capital			
Delivered Equipment :			
Overcoating	7,000		
Pressing	14,000		
Heat Treatment and Outgassing	41,000		
In Line Process Control	6,000		
Assembly	2,000		
(*a*) Total for Delivered Equipment	70,000		
(*b*) Installed Equipment		105,000	292
(*c*) Addition to original Building	37,500		
(*d*) Installed Addition to original Building		45,000	125
(*e*) Total installed Cost ($b + d$)		150,000	417
(*f*) Engineering and Design		45,000	125
(*g*) Preoperational Cost (Six Months Running with Natural Uranium Coated Particles and only Two Weeks Graphite Block Costs Charged)		82,000	228
Total fixed Capital Cost ($e + f + g$)		277,000	770
Working Capital			
One Month Running Cost including Graphite Blocks		49,000	136
55 kg U^{235} in Form of Coated Particles		301,000	837
Spare Parts (5 % of installed Equipment Cost)		5,000	14
Total Working Capital		355,000	987
Total Investment			
Fixed Capital		277,000	770
Working Capital		355,000	987
TOTAL		632,000	1,757

Recoverable rejects and irrecoverable losses are assumed to be 2 % and 0.2 % respectively.

A prismatic form of core arrangement is envisaged, built up from either pressed or extruded graphite blocks, each block containing about 0.15 kg U^{235}. Economic factors favour extrusion of graphite and this will be assumed in this estimate, but it should not be forgotten that in principle a pressed block would have superior mechanical properties and be less prone to gross defects which might modify the technical position by allowing an increase in service lifetime. It has been assumed that the cost of extruded graphite is £600/tonne including machining, this cost being applied to the unmachined block dimensions.

For this extension of the cost estimate, essentially the same methods have been used as were described in Section 3.1. The capital investment summary shown in Table 7 shows that the total of fixed and working capital is £632,000 ($1,750,000). This figure is in addition to that given in Table 4 so that the overall capital investment to make coated particles and consolidate them into fuel elements is about £1.75M ($5,000,000). Table 8 gives the breakdown of costs for the operations required to convert coated particles into fuel elements.

The overall breakdown of costs for the conversion from enriched UF_6 to fuel elements (i.e., Tables 5 and 8 combined) is given in Table 9. It appears that the total fabrication cost (including thorium and graphite but excluding enriched uranium) is about :

£940/kg U^{235} ($2,600/kg U^{235})

or £85/kg heavy metal ($235).

TABLE 8. — Consolidation — Cartridge Route — Breakdown of Costs
(1,110 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10 starting from coated particles)

Item	Cost		
	£/kg U^{235}	$/kg U^{235}	% Total Fabrication Cost (Excluding Coated Particle Cost)
Graphite Blocks	290	806.4	62.4
Graphite Matrix Powder	18.9	52.5	4.1
Overcoating (not including Graphite Matrix Powder)	24.8	68.9	5.3
Pressing	24.3	67.6	5.2
Heat Treatment and Outgassing	41	114	8.8
In Line Process Control	8.3	23.1	1.8
Assembly	7.0	19.5	1.5
Irrecoverable Losses	11.2	31.2	2.4
Recovery of Rejects	16.4	45.6	3.5
Financial Charges	23.1	64.2	5.0
TOTAL	465	1,293	100.0

3.5.3. — *Loose Coated Particle Route.*

The use of loose coated particles loaded directly into a fuel element will lead to a lower fabrication cost than by preforming the coated particles into a matrix. It is assumed that loose coated particles are incorporated into blocks of similar section to those envisaged previously amenable to cheap graphite production. The wisdom of not using an exotic graphite manufacturing process is already evident from Table 8. With this simplified design the total fabrication cost reduces from £940/kg U^{235} ($2,600) to about £770/kg U^{235} ($2,100) or £70/kg heavy metal ($195).

3.5.4. — *Discussion of Costs of Consolidation and Assembly.*

It is seen from Table 8 that the graphite blocks for the prismatic core arrangement dominate the cost of consolidation. This is obvious when it is realised that a C : U^{235} atomic ratio of about 6,000 is desirable [3]. Even were all the carbon atoms purchased as raw graphite capable of being used as moderator the cost could not reduce below about £150/kg U^{235}. Efficient use of graphite should therefore be one of the dominant design features, remembering that *every piece machined off and every hole drilled cost money.*

The savings to be effected by the use of loose (or nearly loose) particles are significant though technical aspects, such as particle failure in service, heat transfer from a packed bed and safety in handling within the Reactor, must dominate this choice. Strong points in favour of loose coated particles are also the minimisation of mechanical damage to the coating, the high fuel density achieved as well as reduction in reprocessing cost.

4. — Alternative Fuel Cycles

In this section brief mention only will be made of alternative fuel cycles of interest to high temperature gas cooled reactors. Obviously many extremely long term factors affect the ultimate choice, in particular the supply of plutonium and its utilisation in other types of reactor has a profound effect.

Initial work within the Dragon Project was concentrated on the U^{235}/Th once-through cycle [10]. Other fuel cycles which deserve attention are :

— U^{235}/U^{233}/Th reprocessing cycle.
— Pu/Th once-through cycle.
— Pu/natural or depleted uranium once-through cycle.
— Combination of the above cycles as feed and breed systems.

The U^{235}/U^{233}/Th cycle is discussed in a separate paper [3]. We consider that this cycle is not likely to come into use until a considerable reactor programme has been mounted and commissioned, i.e., at least five years after the commencement of the first HTR power reactor operation. Since the larger the reprocessing capacity the lower the fabrication cost, this time delay may be even greater if the once-through cycle is shown to be adequately cheap. However it must be conceded that a fuel

conservation policy would introduce reprocessing as early as possible so as to gain experience.

The Pu/Th once-through cycle is also covered in a companion paper at this Symposium [4]. It suffers from the fact that the burn-up of plutonium is not matched by the conversion of Th^{232} to U^{233}. As a consequence the fuel would have to be discarded with a relatively low fifa suggesting that a feed and breed system would be more appropriate.

The physics and fuel cycle costs of the Pu/natural uranium system are also discussed in reference 4. From the fabrication point of view, the greatly increased health hazard compared with handling U^{235} will reflect itself in increased building, equipment and maintenance costs.

5. — Conclusions

The fabrication cost of coated particle fuel given in this paper is derived from experience gained in the manufacture of fuel for the Dragon Reactor Experiment.

There is no doubt that before a high temperature reactor programme is launched the fuel cycle and the core design will be optimised. Nevertheless as long as these reactors continue to use graphite as moderator and coated particle fuel for the primary containment of fission products, the relative influence of the various components of the cost will probably remain as given in Table 9.

Table 9. — High Temperature Reactor Fuel Production Breakdown of Total Fabrication Cost (1,110 kg U^{235}/annum output, thorium/U^{235} atomic ratio N = 10)

	£/kg U^{235}	% Total Fabrication Cost		$/kg U^{235}
Machined Graphite Blocks	290	31		806.4
Conversion from UF_6 to UO_2	100	10.6	50 %	278
Thoria and Carbon (for Particle Making)	59.4	6.4		165
Graphite Matrix Powder	18.9	2.0		52.5
Irrecoverable Losses (1 % + 0.2 %)	62.2	6.6	10.6 %	173
Recovery of Reject (5 % + 2 %)	37.9	4.0		105.4
Weighing	5.5	0.6		15.3
Kernel Preparation	32.4	3.5	21.5 %	90
Reaction Sintering	48.1	5.1		133.8
Coating	114.8	12.3		319
Overcoating	24.8	2.6		68.9
Pressing	24.3	2.6		67.6
Heat Treatment and Outgassing	41	4.4	11.2 %	114
In Line Process Control	8.3	0.9		23.1
Fuel Element Assembly	7	0.7		19.5
Financial Charges	62.4	6.7		173.5
Total	937	100.0		2,605

Fifty per cent of the total fabrication cost is incurred in purchasing the raw materials in a form suitable for use, 33 % being the cost of graphite alone. Consideration should be given to re-use of the graphite blocks after the burnt fuel has been removed. At present there is insufficient information to enable a realistic estimate as to the viability of this suggestion. However, the properties of highly irradiated graphite might well be a limiting feature.

Some 10 % of the total fabrication cost is due to the conversion from UF_6 to UO_2 and this is obviously an area for further technical study.

Another item of interest is the cost of losses and rejects. The amount of reject will probably not be reduced greatly due to the high standard of quality required for nuclear fuel but as indicated earlier the cost of recovery may well be reduced. The strong influence of the loss on the total cost emphasises the need for good engineering of the line and training of the staff in good housekeeping.

The dominant figure in the coated particle fabrication cost (20 % of the total cost) is the 12 % resulting from coating. As has been said previously, coating is the major process on which the greatest technical effort should be placed, in addition to efforts to eliminate the high cost of the $UF_6 \rightarrow UO_2$ conversion.

The consolidation cost is not likely to be reduced unless the incorporation of loose coated particles is shown to be technically feasible.

About half of the financial charges derive from the U^{235} inventory in the fabrication plant. Whether or not the fabricator has actually to purchase the U^{235} will depend on local conditions. In any case the contribution of the financial charges to the total cost is only about 7 %.

The above discussion shows that the main interest of this cost estimate is to focus development effort on those areas where economies are most important.

ACKNOWLEDGMENT

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FABRICATION VARIABLES, PERFORMANCE AND COST CONSIDERATIONS FOR HTGR COATED-PARTICLE FUELS (1)

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ABSTRACT

Studies involving the development of pyrolytic-carbon-coated uranium-thorium carbide particles and of graphite-matrix fuels containing coated particles have produced a basis for HTGR fuel element designs and for fuel-cycle cost analysis. Irradiation tests combined with thermal and mechanical test evaluations have demonstrated satisfactory fuel element performance for pebble-bed reactor application. More recent work on particle coating and evaluation techniques has led to means for improved control of production variables. Pyrolytic-carbon-coated oxide fuel particles offer promise for reduced costs and satisfactory reactor service. The results presented are generally applicable to prismatic fuel structures as well as to spheres.

1. — INTRODUCTION

Operational requirements for nonpurged graphite-based fuel elements for high-temperature gas-cooled reactor (HTGR) applications are associated primarily with fission-product retention, from the standpoint of both hazards and system maintenance. In a broad sense, this means that all components of the fuel system must be mechanically and chemically stable with respect to the reactor environment during reactor service. The results of extensive development programs [1-3] show that such requirements can be met in helium-cooled systems through the use of pyrolytic-carbon-coated fuel particles dispersed in graphite matrices.

Important factors in regard to long-range acceptance of this fuel concept for advanced central-station power production are initial fabrication costs and refabrication costs, with the latter being significant in fuel-cycle considerations for reactor lifetimes up to 30 years [4]. Thus, it is important that optimization with respect to economic factors and to operational requirements be established through design and evaluation experiments.

It is the purpose of this paper to describe the experimental approach and present the results of a research and development program at the Oak Ridge National

(1) ORNL-report TM-1123, April 1965.

(2) Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

Laboratory designed to provide information on the interrelationships involved in optimization. Major emphasis in this program has been placed on investigations of coated-particle-fueled graphite spheres of the type required for the German pebble-bed reactor (AVR) [5, 6,]. A schematic drawing of the type of fuel element to be used in the first loading of the AVR is shown in Fig. 1, and some reference conditions for this application are listed in Table 1. (Requirements for abrasion resistance and impact resistance that are unique to pebble-bed concepts are discussed in the second section of this paper titled « Fueled Graphite Spheres. »). A broad range of variables has been examined, and the results are generally applicable to prismatic fuel-element designs for both purged and nonpurged HTGR systems, as well as to nonpurged systems that use spherical fuel elements.

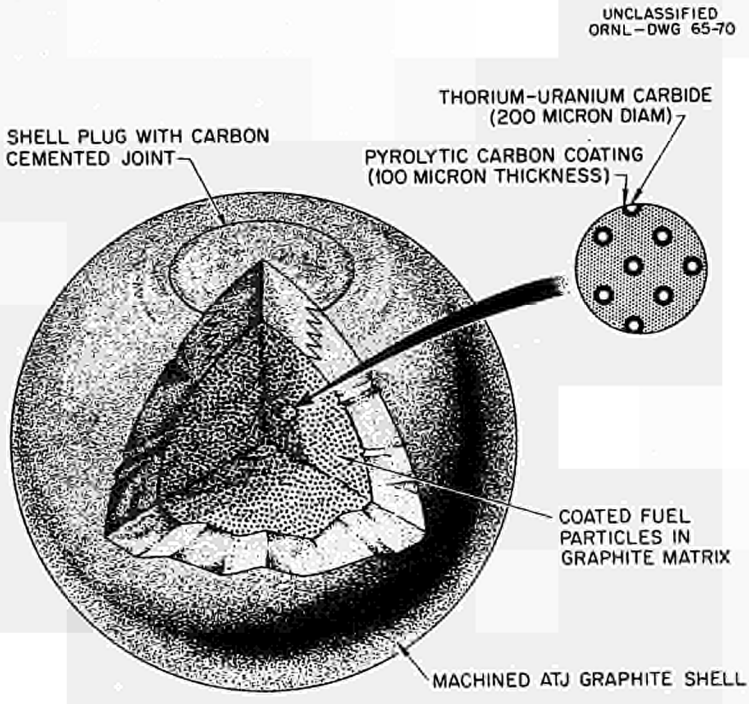

FIG. 1. — Schematic Drawing of Fuel Element to be Used in First Loading of AVR Reactor.

The topics covered in this paper — all with direct or indirect reference to cost considerations — are listed below :

1. Effect of coating-deposition parameters on the properties and performance of pyrolytic-carbon-coated fuel particles.
2. Preparation and performance of pyrolytic-carbon-coated oxide fuel particles.

3. Effects of fabrication variables on the performance of fueled-graphite spheres for pebble-bed reactor applications.

TABLE 1. — Reference Conditions for AVR Fuel Spheres Used in ORNL Development Program

Sphere dimensions, mm	$60.0 \pm \frac{0.0}{0.2}$
Fuel loading, wt % heavy metal	5
Thorium-to-uranium ratio	7 : 1
Burnup, at. % heavy metal	7 to 8
Surface temperature, °C	900 to 950
Power per sphere, kw	
Average	1.4
Maximum	2.1
Fission-product release, R/B for ^{88}Kr	$\leq 10^{-4}$

2. — COATED-PARTICLE STUDIES

The demonstration of a high level of irradiation performance for coated particles with certain general features [1, 2] set the stage for the systematic studies described in this section. The coated particles investigated most extensively consist of spheroidal carbide or oxide particles, approximately 200 μ in diameter, coated with approximately 100 μ of pyrolytic carbon by the fluidized-bed technique.

These studies were designed to generate information that will provide a better understanding of the specific properties desired and to establish processing conditions for achieving these properties economically. The incentive for close examination of economical processing conditions was provided by the cost analysis of Lotts *et al.* [7] for remote refabrication of HTGR fuel elements. According to this analysis the coating operation accounts for 31 to 41 % of the cost for plant capacities of 60 to 3,700 kg of heavy metal per day.

2.1. — EFFECT OF DEPOSITION CONDITIONS ON COATING PROPERTIES.

Techniques have been developed for fluidized-bed coating of carbide and oxide fuel particles with carbon formed by the pyrolysis of methane or acetylene under precisely controlled conditions. The dominant variables affecting coating structure are deposition temperature and the rate at which the coating is applied, although it is recognized that any change in any part of a fluidized-bed coating system affects the properties of the product.

2.1.1. — *Coatings Deposited from Methane.*

Coating experiments involving methane were carried out at eight temperature levels corresponding to 100° C increments between 1300 and 2000° C. At each

temperature the total gas-flow rate and charge size were fixed and the same coating apparatus was used so that the only variable was the methane flow rate and the associated methane partial pressure. In all experiments the system was at atmospheric pressure and helium was used as the diluent or inert carrier gas. The range of methane flow rates studied was 0.0167 to 2.53 $cm^3\ min^{-1}\ cm^{-2}$ (average deposition rates of 2 to 140μ/hr). Details of the experimental conditions and the evaluation techniques employed are described elsewhere [8].

The results of this study are presented graphically in Fig. 2 and show clearly the effect of controllable deposition variables on the density, anisotropy, and crystallite size of the coatings. More recent work shows that the trends indicated in Fig. 2 extend to methane flow rates of 3.3 $cm^3\ min^{-1}\ cm^{-2}$ (deposition rates to 350 μ/hr). These results demonstrate that the properties of coatings deposited from methane can be controlled and that coatings having selected characteristics may be produced for optimization studies with respect to performance and the economics of the use of high coating deposition rates.

2.1.2. — *Porous Inner Layers Deposited from Acetylene.*

Multiple-layer pyrolytic-carbon coatings with an inner porous layer (50 % porosity or more) performed remarkably well under severe irradiation test conditions, as described in the next section. Porous layers of this type are deposited at very high rates from acetylene at relatively low temperatures (1000-1100° C) [1, 2, 9] and are therefore attractive, in principle, from a cost standpoint as well. Adequate control of the high-deposition-rate material and high coating efficiencies must be realized in production-scale operation if full exploitation of this concept is to be realized.

2.2. — PYROLYTIC-CARBON-COATED OXIDE FUEL PARTICLES.

Retention of the structural integrity of pyrolytic-carbon coatings on carbide fuel particles under severe irradiation conditions led to serious consideration of utilizing carbon-coated oxide fuel particles for HTGR applications. A matter of possible immediate concern is the basic thermodynamic instability of oxide fuel compounds and carbon under both fabrication and operational conditions; however, this concern may not have basis in practice if it can be demonstrated that the low-permeability coatings can be applied satisfactorily and that they will retain their integrity so as to withstand the equilibrium partial pressures of the reaction-product gases.

There appears to be considerable economic incentive to utilize oxide particles and thus avoid the costly steps in preparing carbides. The conversion of particles from oxide to carbide is difficult to achieve without damage to the particles from sticking or sintering. The handling problems associated with the highly reactive thorium-rich carbide materials also increase costs. Recent pilot-scale preparation of oxide microspheres by the sol-gel technique at the Oak Ridge National Laboratory [10]

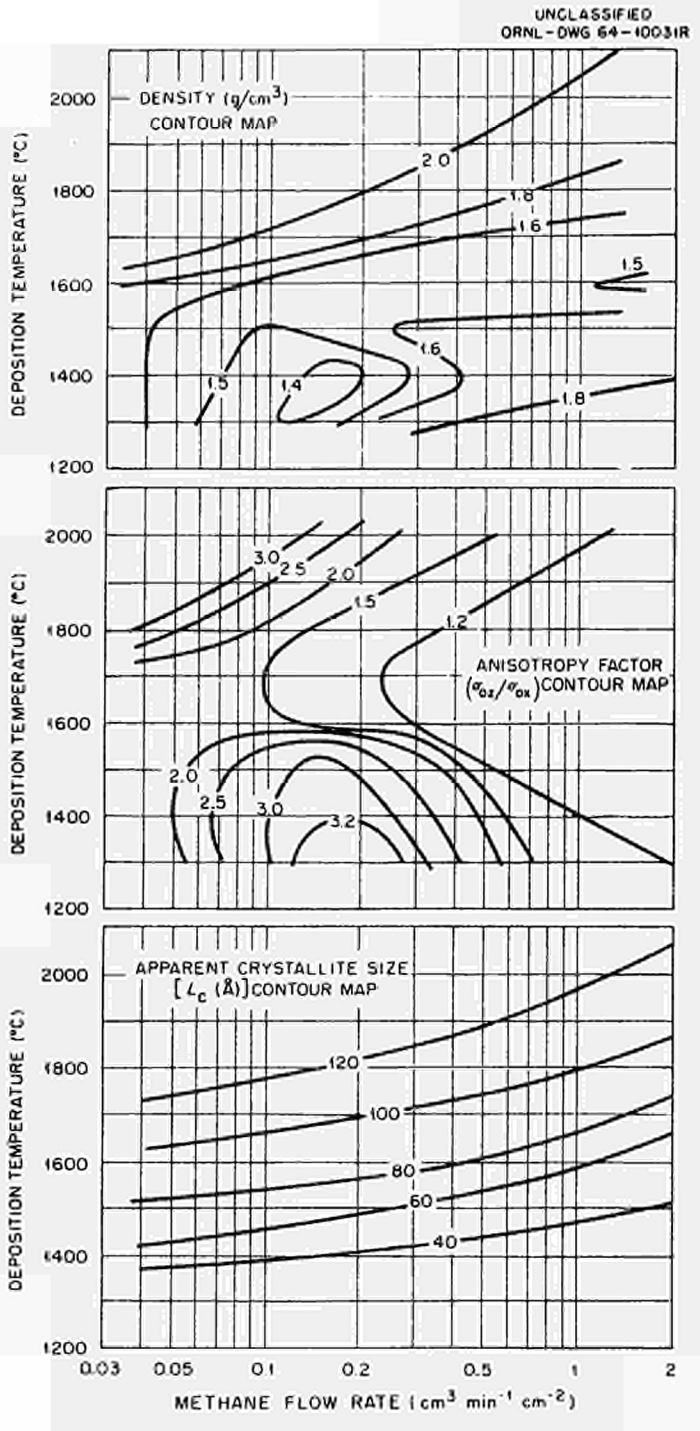

FIG. 2. — Effect of Methane Flow Rate and Temperature on the Properties of Pyrolytic-Carbon Coatings Deposited from Methane on 200-μ-diam Uranium Carbide Particles.

has provided further incentive to exploit this concept. Nearly theoretically dense, spherical $(Th,U)O_2$ particles can be produced by this process in a narrow range of sizes with compositions from ThO_2 to UO_2. Temperatures of only about 1200° C are required for final densification. It is estimated that reductions in fuel fabrication costs of greater than 15 % can be achieved if oxide, rather than carbide, particles are used [7].

In view of these considerations, a program was initiated at ORNL for the coating of oxide fuel particles and for the determination of the thermal and irradiation stability of coated oxide particles under conditions likely to be encountered by HTGR fuel elements during fabrication and operation. The results of the irradiation tests are presented in the next section on « Irradiation Test Results. ».

2.2.1. — *Coating Conditions.*

Carbon coatings were applied to fully dense sol-gel ThO_2 and $(Th,U)O_2$ particles and to conventionally prepared (sintered) UO_2 particles from methane and acetylene in fluidized beds. The UO_2 particles investigated were of low bulk density (80-85% of theoretical), and they had very rough surfaces and a poor degree of sphericity. Before these UO_2 particles were coated with pyrolytic carbon, approximately 10 to 15 μ of powdered graphite was applied to them by a wet-spray technique to provide a more regular surface for coating. The coatings deposited from methane were applied at 1400 and 1800° C; the relatively more porous ones deposited from acetylene were applied at 900 and 1100° C. Individual batches with coatings ranging in average thickness from about 30 to 155 μ on 150- to 250-μ-diam particles were prepared for the out-of-reactor thermal stability studies.

2.2.2. — *Thermal Stability Tests.*

Heat treatments were carried out in vacuum (10^{-4} — 10^{-5} torr) at 1100° C for 1000 hr, at 1400° C for 200 hr, at 1800° C for 4 hr, at 2000° C for 50 hr, and at 2400 to 2700° C for 2 min. One atmosphere of argon was used for thermal treatments at 2000° C for 2 to 100 hr and at 2200° C for 2 hr. A total of 22 different batches of coated particles was used in these tests, and evaluation was in terms of microradiography, metallography, and x-ray diffraction [11].

No evidence of reaction between coatings and particles or of heavy-metal migration was observed in any of the samples in the as-deposited condition and after heat treatments at 1100 and 1400° C. At 1800° C and higher temperatures, rupture of the coatings and subsequent conversion of the particles to carbide was observed in some cases, but not always, for samples with relatively thin coatings (20 to 60 μ deposited from acetylene and 30 to 60 μ deposited from methane). These results indicate that coating thickness becomes an increasingly important factor with increasing heat-treatment temperature. When failure occured it was more or less catastrophic in the form of severely ruptured coatings. No migration of heavy metal was ever observed in sound coatings, regardless of the time and temperature employed. Thus adequately thick coatings (greater than about 60 μ for the extreme high tempe-

ratures) are apparently required to withstand the equilibrium CO pressures developed during elevated temperature exposures. It was noted, however, that two batches of particles with coatings deposited from methane to average thicknesses of 43 and 50 μ survived treatments at 2000° C for 50 hr and at 2200° C for 2 hr with no detectable damage. The role of coating structure is being investigated.

2.3. — Irradiation Test Results.

Significant results of irradiation tests on unsupported coated particles at ORNL have been described in detail [1, 2, 11-13], except for the results of the more recent tests involving coated oxide particles. The test facilities have included static capsules inserted in the Low-Intensity Test Reactor (LITR) and the Materials Test Reactor (MTR), sweep capsules in the Oak Ridge Research Reactor (ORR), and, for coated oxide particles only, the loop No. 1 facility in the ORR.

The results of static tests have generally corroborated those of sweep tests. Results from sweep facilities are rather emphasized in this discussion because in these tests a continuous account of the behavior of the test material is provided by analysis of the helium gas that is passed through the capsule. This gas is monitored continuously for total activity and sampled periodically for fission-gas analysis. Similar information is available, of course, from the circulating helium gas in loop tests, as discussed below under the heading "Coated Oxide Particles." The test conditions and results are summarized below and information on the higher performance tests involving multilayer coatings is listed in Table 2. Characterization of the coatings on the ORNL-prepared particles listed in Table 2 is given in Table 3.

Table 2. — Irradiation Test Conditions and Fission-Gas Release Data for Unsupported Coated Fuel Particles with Multilayer Pyrolytic-Carbon Coatings

Experiment	Sample Designation	Type of Fuel Particle	Coating Structure	Test Temperature (°C)	Burnup (at. % heavy metal)	R/B for Kr^{88}
B9-15	GA-309	UC_2	Triplex [a]	1,370	18.7	4.0×10^{-5}
B9-16	NCC-216	UC_2	Duplex [b]	1,370	18.8	1.0×10^{-5}
B9-17	OR-138	UC_2	OR-2D [c]	1,370	14.9	1.1×10^{-5}
B9-19	OR-206	(Th, U)O_2 [d]	OR-2D [c]	1,200	0.6	2.3×10^{-6}
B9-20	OR-201	UO_2	OR-2D [c]	1,370	4.2 [e]	1.5×10^{-6}
A9-2	OR-298	UO_2	OR-2D [c]	1,400	8 [f]	2.0×10^{-7}
Loop 1-14	OR-206	(Th, U)O_2 [d]	OR-2D [c]	1,370	2.7 [g]	2.5×10^{-5}

[a] See refs. 1, 2 and 9.
[b] See R. A. Reuter, Duplex Carbon-Coated Fuel Particles, *Nucl. Sci. Eng.*, 20(2) : 219-226 (1964).
[c] Two-layer coatings described in Table 3; properties can be determined by reference to Fig. 2.
[d] Particles contain 8 wt % UO_2 and were prepared by the ORNL sol-gel process.
[e] Test terminated due to thermocouple failure.
[f] Test still in progress.
[g] Test still in progress; about 9 months of irradiation has accumulated at conditions indicated.

TABLE 3. — Characterization of OR-2D Coatings Deposited from Methane and Listed in Table 2.

Sample Designation	OR-138	OR-201	OR-206	OR-298
Type of fuel particle	UC_2	UO_2	$(Th, U)O_2$	UO_2
Average fuel-particle diameter, μ	186	229	206	302
Inner layer				
Coating temperature, °C	1,400	1,400	1,400	1,400
CH_4 flow rate, $cm^3\ min^{-1}\ cm^{-2}$	0.17	2.8	0.83	0.16
Coating thickness, μ	40	40	35	70
Outer layer				
Coating temperature, °C	1,700	1,800	1,800	1,900
CH_4 flow rate, $cm^3\ min^{-1}\ cm^{-2}$	0.17	0.27	0.17	0.10
Coating thickness, μ	72	80	63	65

2.3.1. — *Coated Carbide Particles.*

The following conclusions were drawn from extensive testing of coated, high-density UC_2 and $(Th, U)C_2$ fuel particles with nominal particle diameters of 200 μ : (1) multilayer coatings are superior to the monolithic coatings tested; (2) the performance of monolithic coatings with average thicknesses of 50 to 75 μ is substantially inferior to that for 100-μ-thick coatings; (3) spheroidal particles generally perform better than irregularly shaped particles with sharp edges and protrusions; (4) in experiments in which no coatings have ruptured during irradiation, the fission-gas release, expressed as the ratio of the release rate to the birth rate (R/B) for ^{88}Kr, has been of the order of 10^{-5} to 10^{-6} and can be accounted for by the uranium contamination in the coatings as determined before irradiation by alpha-counting; and (5) the performance of coated $(Th,U)C_2$ particles with Th : U ratios to 2.2. : 1 has been comparable to that of similarly coated UC_2 particles.

Excellent overall performance has been demonstrated in tests on coated carbide particles with the so-called "triplex" coatings having porous inner layers [9]. After a burnup of 18.7 at. % heavy metal at 1370° C, none of the coatings had ruptured and there was no evidence of coating damage; only a slight swelling of the fuel particles into the porous inner layers was detected. Similar performance from the standpoint of fission-gas release has been exhibited by two-layer coatings without the very porous inner layer, as shown in Table 2. In these cases, however, the innermost coating layers were more or less severely damaged by the usual wedge-shaped fractures and in-flow of the carbide fuel, but none of these intrusions had proceeded past the physical discontinuity at the interfaces between the two layers at the burnups indicated.

2.3.2. — *Coated Oxide Particles.*

The highest burnup demonstrated, at this writing, for loose coated oxide particles is 8 at. % heavy metal at 1400° C in a sweep capsule (experiment A9-2). This

test has operated for about 7 weeks and, by comparison with results from other tests in this type of facility, the fission-gas release has been constant and very low. Thermocouple failure occasioned termination of a previous sweep capsule test (experiment B9-20) on coated UO_2 particles after a burnup of 4.2 at. % heavy metal at 1370° C. None of the coatings was ruptured in this test as the result of irradiation, and metallographic examination revealed only minor damage by wedge-shaped fractures, none of which extended beyond one-half the thickness of the inner coating.

Sol-gel $(Th,U)O_2$ particles containing 8 % UO_2 and coated with a two-layer coating, as described in Table 3, have been irradiated at 1200° C to a short burnup in experiment B9-19 and for about 9 months to a burnup of 2.7 at. % heavy metal in the ORR loop No. 1 facility [14, 15] at 1370° C. The consistently satisfactory performance of this material in the loop test lends encouragement to the possibility that relatively economical coated particles of this type will retain their integrity for extended periods of time at fuel temperatures of current interest in the circulating environment of HTGR systems.

3. — Fueled-Graphite Spheres

As pointed out in the introduction to this paper, major emphasis in the HTGR fuel element test program at ORNL has been placed on 6-cm-diam fueled-graphite spheres in view of the AVR requirements listed in Table 1. Some comments on the general configurations considered, the fabrication techniques examined, and the effects of fabrication variables on the mechanical properties and irradiation test performance are presented in this section.

3.1. — General Configurations Considered.

The fuel spheres in a pebble-bed reactor are subjected to impact loading and abrasive action during reactor operation, and therefore it is mandatory that the coated fuel particles be protected by an unfueled graphite shell. In the absence of such a shell, the coatings on particles at the surface of the sphere could be ruptured and the fission-product retention criteria would not be met.

A major design consideration involves the choice between all molded concepts, i.e., configurations in which the unfueled shell is molded and baked, or "hot pressed," as an integral part of the final product, and so-called "machined-shell" concepts in which the unfueled shell is initially machined from fully graphitized material and the graphite matrix fuel region is inserted subsequently by one technique or another. Other important design considerations include (1) specification of the shell thickness adequate for mechanical property requirements but not so thick as to give rise to inordinately high central temperatures at maximum sphere power density, (2) the type of graphite formulation or commercially available graphite to be used for the shell, and (3) relative fabrication costs associated with suitable configurational designs.

Calculated center temperatures for various shell thicknesses and the effect of a 0.015-in. (380 μ) shell-core gap are shown in Table 4 for the AVR design criteria used in the ORNL program. These results indicate strongly that, in the light of the coated-particle irradiation performance data available, shells thicker than 0.8 cm would not be desirable if shell-core gaps of 0.015 in. were formed.

TABLE 4. — Calculated Center Temperatures and Fuel Concentrations for 6-cm-diam Spheres as a Function of Unfueled Shell Thickness

Shell Thickness (cm)	Concentration of Coated Particles [a] (vol. %)	Interparticle Spacing [b] (μ)	Calculated Center Temperature [c] (°C)	
			No. Gap	0.015-in. (380-μ) Gap
0	6.6	400	1,100	
0.6	12.9	230	1,190	1,390
0.8	16.7	180	1,240	1,470
1.0	22.2	130	1,285	1,560
1.2	30.5	75	1,350	1,670

[a] For 5 wt % heavy metal fuel loading as 200-μ particles with 100-μ coatings.
[b] Closest approach for ideal cubic array.
[c] For a power of 2.1 kW per sphere and a surface temperature of 900° C; thermal conductivities of fueled region and unfueled shell assumed to be 0.14 and 0.33 w/cm °C, respectively, and gap assumed to contain helium.

3.2. — FABRICATION TECHNIQUES.

Matrix formulations for spherical graphite fuel elements containing coated carbide fuel particles require graphite flour, rather than coke, as the filler material, since heating to graphitization temperatures would damage the coated particles. Fabrication techniques applicable to the fueled region of this type of element thus include cold pressing followed by binder curing and baking, warm pressing and baking, "hot pressing" (see, for example, ref. 9), and mold injection and baking [16].

For cold- and warm-pressing techniques, particularly, it is clear that for the high fuel loadings considerable care must be exercised during pressing to prevent damage to the coatings This effect is shown in Fig. 3 for isostatically pressed (3,000 to 35,000 psi) formulations containing 200-μ-diam particles with 100-μ-thick monolithic laminar-type coatings as a function of fuel loading (6 to 23 vol % coated particles). These data show that cold pressing pressures as high as 6,000 psi are acceptable for loadings up to 21 vol %.

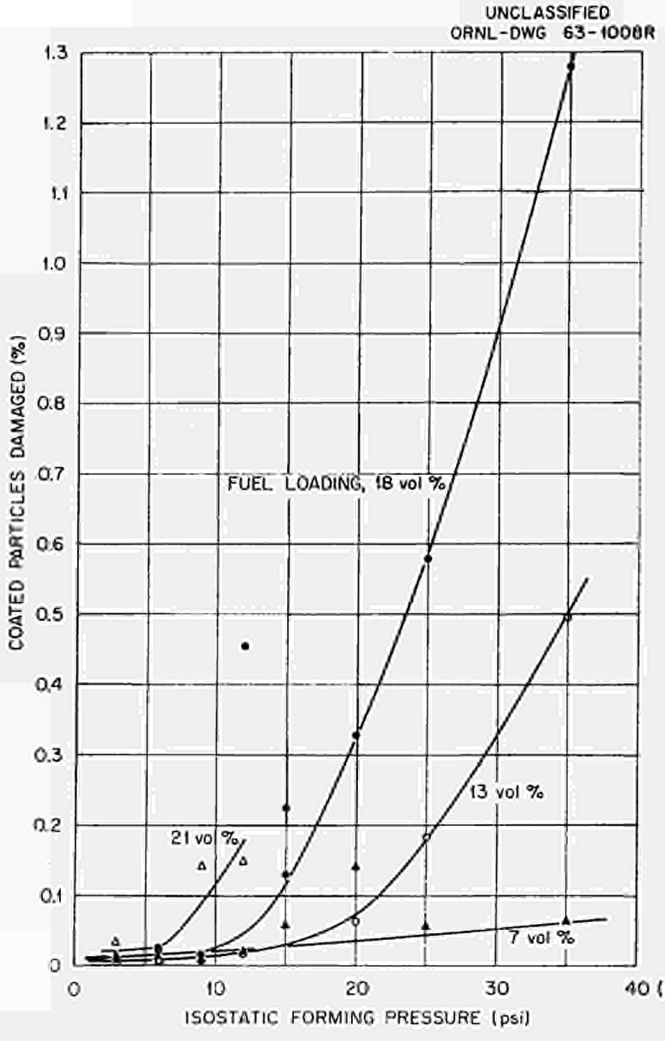

FIG. 3. — Effect of Isostatic Forming Pressure and Fuel Concentration on Damage to Laminar Pyrolytic-Carbon-Coated Particles in Graphite Matrices.

3.3. — EFFECTS OF FABRICATION VARIABLES ON MECHANICAL PROPERTIES.

The mechanical properties of greatest importance for pebble-bed HTGR systems are impact and abrasion resistance. Although abrasion resistance is difficult to define, it was established in the AVR program at Julich that fully graphitized, machined, nuclear-grade material (Siemens-Plania Grade ALSw-AMT) exhibited adequate abrasion resistance in simulated full-scale pebble-bed tests. This consideration and the results of the impact tests described below, as well as cost studies for fabrication of molded versus machined-shell spheres, led to an early decision to place emphasis on the machined-shell concept in the program.

An impact-test program designed to assist in the final design of an AVR-type fuel sphere with a premolded spherical fuel insert cemented in a machined shell with a threaded plug was undertaken. Three related courses were pursued : (1) evaluation of commercially available graphite, (2) determination of the minimum shell thickness required, and (3) study of the effects of insert-shell gaps, which could be present initially or develop during reactor operation. The specification provided that, after 10 thermal cycles in vacuum between room temperature and 1000° C, each 6-cm-diam fuel sphere must be able to sustain 50 drops from a height of 4 meters onto a close-packed bed of 6-cm-diam solid graphite spheres three layers deep. The details of this program have been described [17].

The role of the type of graphite, i.e., extruded versus molded, was determined by dropping solid spheres representative of commercial products onto a steel plate. The general superiority of the molded graphite is shown in Fig. 4, which also shows an interesting correlation between slow-strain-rate crushing strength and impact resistance. Use has been made of this correlation in postirradiation evaluations, as discussed below under "Effects of Fabrication Variables on Irradiation Test Performance."

The effect of fabrication technique and thermal cycling on the impact behavior of fueled spheres is shown in Table 5. Fabrication techniques for preparation of these spheres are tabulated in Table 6 in a later section of this paper dealing with irradiation test results. The effect of radial gap width on impact properties is shown in Fig. 5.

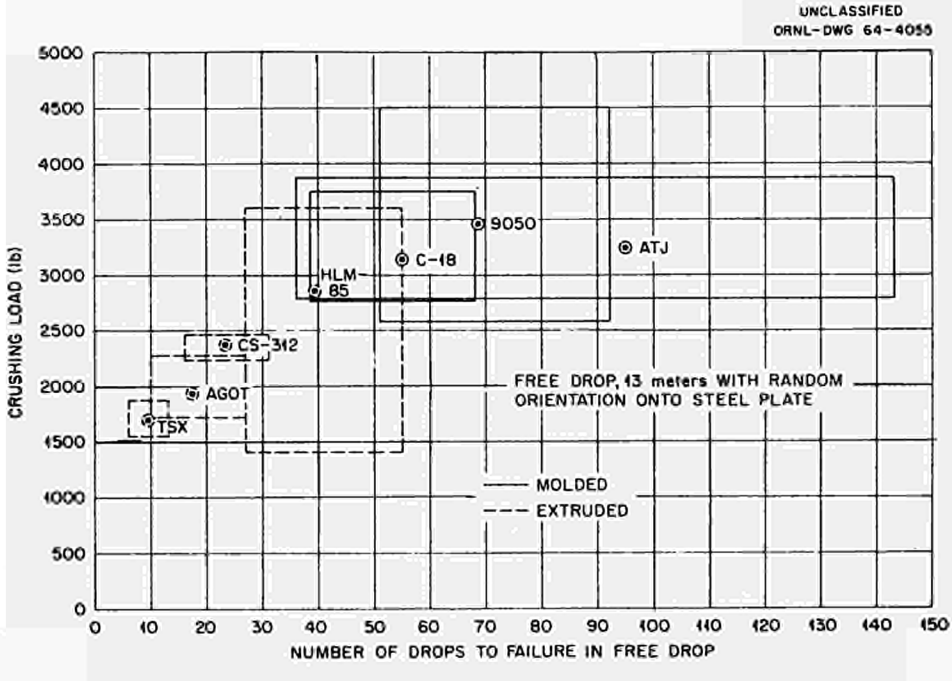

FIG. 4. — Relationship Between Free-Drop Resistance and Crushing Strength of Seven Types of Graphites in the Form of 1½-in.-diam Solid Spheres.

TABLE 5. — Effect of Fabrication Technique and Thermal Cycling on Impact Resistance of 6-cm-diam Fueled-Graphite Spheres

Shell	Specimen	Thermally Cycled	Number of Specimens	Number of Drops Onto Bed of Spheres[a]	Subsequent Number of Drops to Failure from 4 Meters Onto Steel Plate		
					Minimum	Average	Maximum
Machined	GA-VS15	No	2	75	1	1	1
	GA-VS16	No	2	75	1	1	1
	NCL-VS2	No	2	75	1	3	5
	NCL-VS2	Yes	2	75	1	2	3
Molded	3M-VS15	No	3	75	76	95	114
	3M-VS15	Yes	3	75	75	90	105
	3M-VS17	No	2	75	109	163	217
	3M-VS17	Yes	3	75	91	132	182
	3M-VS18	No	3	75	254	266	279
	3M-VS18	Yes	2[b]	75	111	176	240

[a] Without failure.
[b] A third specimen failed on the 16th drop onto the steel plate. The mode of failure strongly suggested poor bonding between core and shell.

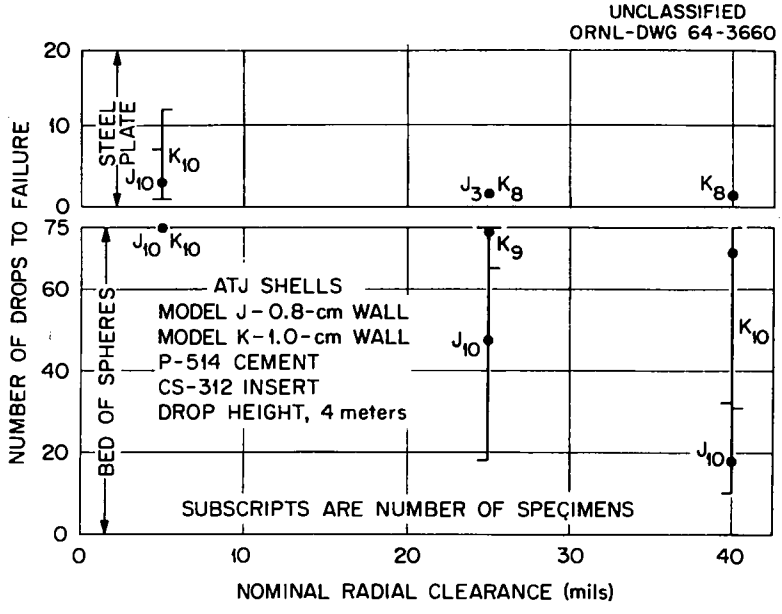

FIG. 5. — Effect of Radial Clearance on Impact Properties of Simulated AVR-Type Fuel Spheres with Machined Steels.

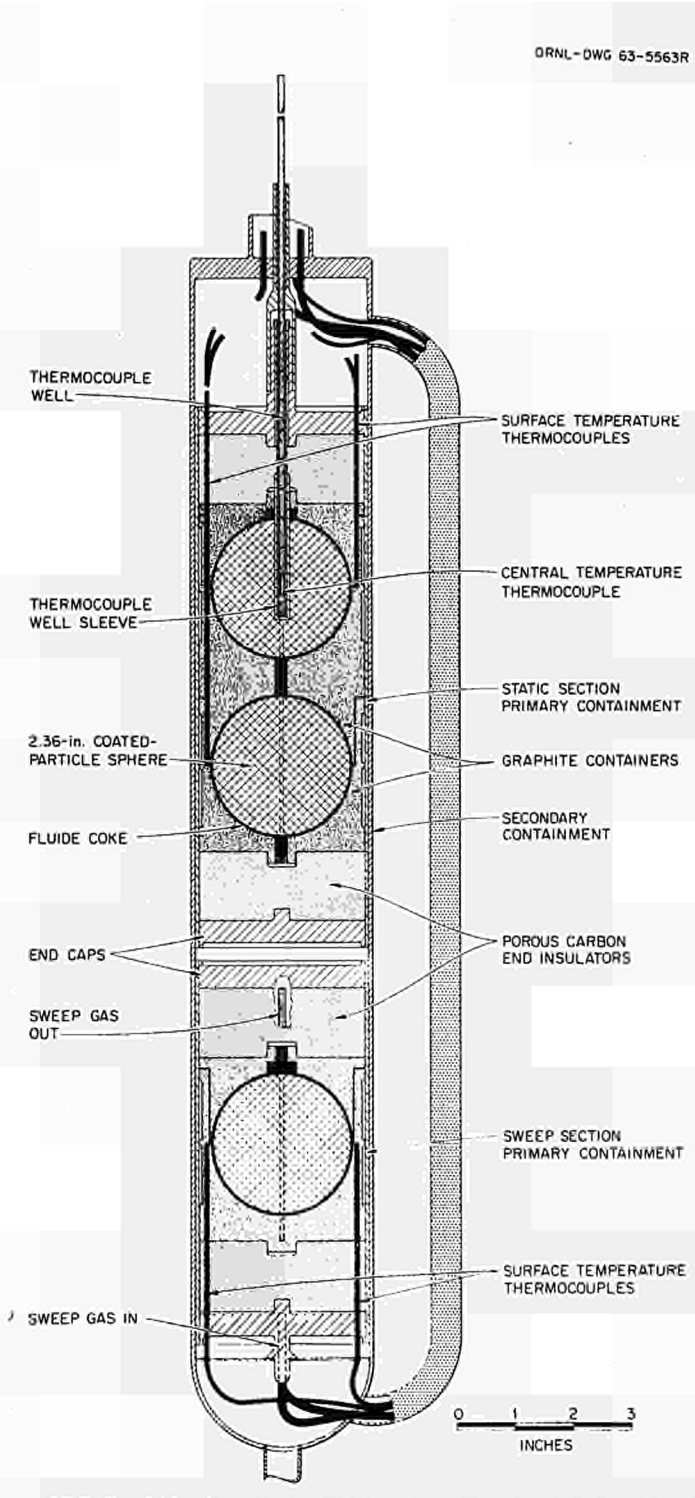

FIG. 6. — ORR Poolside Capsule for Testing 6-cm-diam Fueled Graphite Spheres.

On the basis of these studies, it was concluded that (1) thermal cycling in vacuum has no effect on impact resistance (2) elements with uncemented inserts are clearly inferior to elements with completely cemented inserts, and (3) shells with 0.8-cm-thick walls are satisfactory for a nominal radial clearance between insert and shell of 0.005 to 0.010 in. (130 to 250 μ), but 1.0- to 1.2-cm-thick shells would be required for clearances of 0.020 to 0.025 in. (510 to 630 μ). Limited data indicate that all-molded elements are superior to machined-shell elements, although they lose some impact resistance upon thermal cycling in vacuum.

3.4. — Effects of Fabrication Variables on Irradiation Test Performance.

3.4.1. — *Irradiation Test Performance, Activity Release, and Mechanical Properties.*

Evaluation of a reactor fuel element must take into account irradiation test performance. Although it is desirable to test in actual power reactor service, no suitable facility yet exists for in-service testing of HTGR fuels. The irradiation capsule and loop tests now employed in evaluating spherical fuel elements offer advantages for instrumentation and control and for measurement of fission-gas release as a function of irradiation experience.

Most capsule irradiations have been conducted in the ORR poolside facility, which provides the reference conditions, including fission power density, temperature, and a sweep atmosphere of helium. Other capsules in core positions have been used to obtain high burnup, and a recirculating loop has provided more realistic helium coolant conditions for two experiments. The design of the poolside irradiation capsule is shown in the schematic diagram of Fig. 6. These irradiation test facilities have been described in detail [14].

ORNL hot cells are used for careful examination of the spheres following irradiation. The examinations include a gross gamma scan, visual and photographic observations, dimensional measurements, crushing and impact strength measurements, burnup determinations, metallography, and radiochemical analyses of parts from the graphite structures. The apparatus and techniques used for the examinations were described in a recent publication [18].

Operating conditions and fission-gas release during irradiation for the spheres used in several tests are given in Table 6. It is notable that only one sphere showed a ratio of fission-gas release rate to birth rate, R/B, significantly in excess of 10^{-3}. This sphere, GA-VSII-2, fabricated as shown in Table 6, showed evidence of excessively high temperatures in the matrix and probably operated at a much higher temperature than was estimated because of lack of bonding and the development of a gap, which was observed in postirradiation examination. Relative fractional release of noble fission gases is illustrated in Fig. 7, which shows little difference between the molded and machined shells. A somewhat wider variation is apparent for different elements having machined shells. However, this probably indicates variation in the effectiveness of bonding between the matrix and shell. The shell bonding difficulty may not be inherent in the design, since these were all experimental

TABLE 6. — Operating Conditions and Fission-Gas Release Data for 6-cm-diam ORR-Irradiated Fuel Spheres Containing Coated Carbide Fuel Particles.

Type of Shell	Sphere	Shell Thickness (cm)	Insert Fabrication	Unfueled Shell	Particle		Burnup (at. % heavy metal)	Operating Time (days)	Average Power Density (w/cm^3)	Average Temperature (°C)		R/B	
					Type	Coating				Surface	Center Estimated	^{88}Kr	^{133}Xe
					Poolside Capsules							× 10^{-4}	× 10^{-4}
Machined	GA-VS11	1.1	Hot pressed at 750° C, baked at 1800° C	Type ATJ	$(U, Th)C_2$	Triplex	3.2	87	30	930	1,370	1 to 43 [a]	1 to 43 [a]
	GA-VS15	0.8	Hot pressed at 750° C, baked at 1800° C	Type ATJ	$(U, Th)C_2$	Triplex	2.0	92	18.6	910	Not determined	2.5	6
	GA-VS16 [b]	0.8	Warm pressed at 100° C, baked at 1800° C	Type ATJ	$(U, Th)C_2$	Triplex	2.3	96	18.6	920	1,370	2	4
	NCL-VS1	1.1	Mold injection baked at 1,400° C	Type ATJ	$(U, Th)C_2$	Duplex	1.5	46 [c]	18.6	920	1,320	2	4
	ORNL-VS3	0.8	Warm pressed at 160° C, baked at 1,000° C	Type ATJ	UC_2	Duplex	1.2	62	16	850	1,180	0.2	0.3
Molded	GA-VS12 [d]	0.63	Warm pressed at 150° C	Hot pressed at 750° C, baked at 1800° C	$(U, Th)C_2$	Triplex	4.3	146	30	920	Not determined	3	2.5
	GA-VS12 [d]	0.63	Warm pressed at 150° C	Hot pressed at 750° C, baked at 1,800° C	$(U, Th)C_2$	Triplex	2.7	87	30	940	Not determined	2	2.5
	3M-VS16 [b]	0.63	Not known	Not known	$(U, Th)C_2$	Laminar	3.0	140	18.6	910	1,270	0.01 to 1 [a]	0.01 to 1 [a]
					Loop 2								
Machined	BA-VS13-1	1.0	Hot pressed at 750° C, baked at 1,800° C	Type ATJ	UC_2	Triplex	2.1	98	17.5	710	910	0.1	0.04
	GA-VS13-2	1.0	Same	Type ATJ	UC_2	Triplex	1.4	98		690	800		
	GA-VS13-3	1.0	Same	Type ATJ	UC_2	Triplex	1.0	98	—	700	750		

[a] R/B increased steadily throughout irradiation. [b] Similar spheres. [c] Test terminated by leak in capsule. [d] Similar spheres.

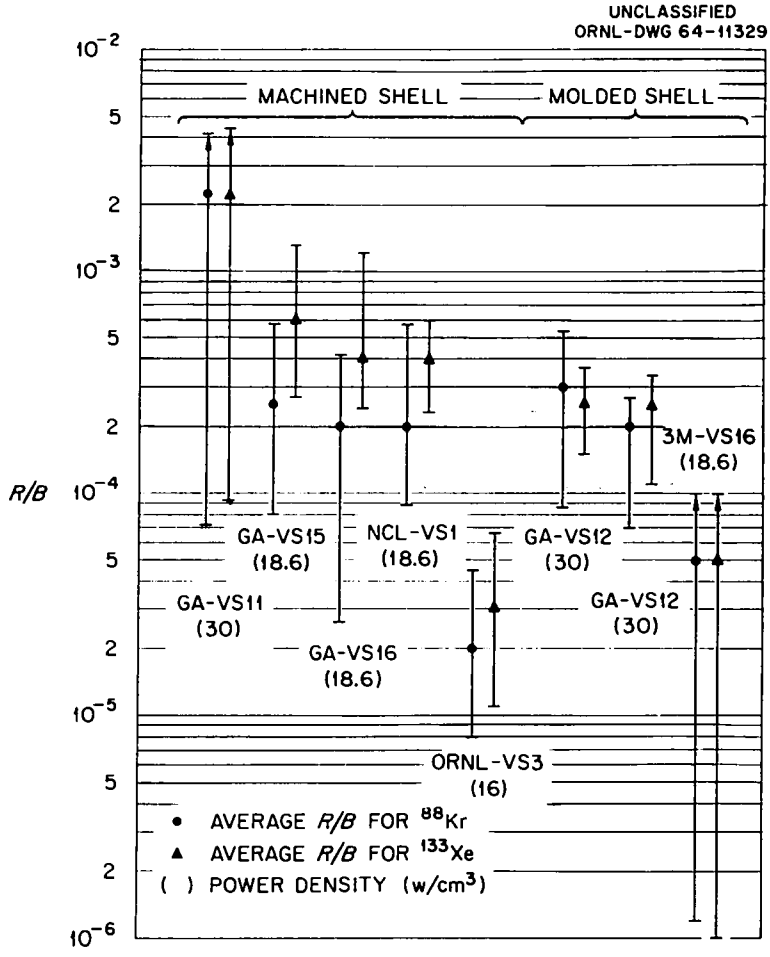

FIG. 7. — R/B Values of ^{88}Kr and ^{133}Xe for 6-cm-diam Fuel Spheres Irradiated in ORR Poolside Facility.

elements, but this does represent an important problem area. Sphere 3M-VS16 showed, at the start, very low fission-gas release but the release increased steadily with burnup. This behavior is attributed to progressive failure of monolithic-laminar coating with increasing burnup (see above).

The tests conducted in ORR loop 2 were of three and four spheres cooled by recirculating helium containing some gaseous impurities compared with the highly purified gas in the sweep capsules. The concentration of hydrogen was allowed to reach more than 300 ppm for limited periods but was below 50 ppm most of the time. Carbon dioxide and carbon monoxide usually were less than 5 ppm. The low fission-gas release rate probably results principally from the low temperature irradiation. There was no indication of carbon mass transfer or other deterioration of these spheres.

TABLE 7. — Effect of Irradiation on Load to Failure in Compression for Fueled-Graphite Spheres

Capsule	Position	Sphere Designation	Shell		Load to Failure (*lb*)	
			Thickness (in.)	Type	Unirradiated	Irradiated
		NCF-S2	0		1,540, 1,550, 1,825	
06Z-6	1	NCF-S2-G				1,400
	2	NCF-S2-K				1,375
	4	NCF-S2-T				1,500
		NCP-SS1	0.43	Machined ZTA	1,075, 1,150, 1,450	
08-6	2	NCP-SS1-4				1,350
	3	NCP-SS1-3				1,300
		GA-S10	0		1,320, 1,380, 1,590	
05Z-6	3	GA-S10-9				1,490
		3M-S4	0		2,760, 2,880, 3,020	
05Z-6	4	3M-S4-11				2,700
		3M-S5	1/16	Molded	3,400, 3,480, 3,620	
06X-6	4	3M-S5-11				2,620
		GA-S9	1/4	Molded	900, 940, 1,100	
08A-6	3	GA-S9-2				1,075
		GA-S8	1/4	Molded	1,080, 1,100, 1,100	
08A-6	4	GA-S8-8				975

		NCF-S6	1/4	Molded	620, 890	
F1-8B-1	2	NCF-S6-1				890
	4	NCF-S6-3				630
	6	NCF-S6-9				560
		SCC-S15	1/8	Molded	Not determined	
F1-8B-2	2	SCC-S15-3				1,350
	7	SCC-S15-5				1,400
		SCC-S16	0.2		Not determined	
F1-8B-3	2	SCC-S16-7		Machined ATJ		1,350
	3	SCC-S16-3		Machined C-18		1,550
	4	SCC-S16-1		Machined AGOT		1,150
	5	SCC-S16-6		Machined CS-312		1,200
	6	SCC-S16-10		Machined ATJ		1,190
	7	SCC-S16-4		Machined C-18		640
	8	SCC-S16-2		Machined AGOT		1,200
		NCF-VS15	0.4	Machined ATJ	800, 960, 1,100	
05-7	2	NCF-VS15-9				1,300
		GA-VS11	0.4	Machined ATJ		
05-7	3	GA-VS11-2				1,450
		GA-VS12	1/4	Molded	2,750	
08-7	2	GA-VS12-3				2,350
	3	GA-VS12-2				2,500
Loop 2, experiment 1		NCF-S4	0		1,360, 1,680, 1,680	
	1	NCF-S4-6				1,510
	2	NCF-S4-4				1,410
	3	NCF-S4-3				1,360

Data from crushing strength measurements are given in Table 7. Again, it is evident that bonding of the matrix and shell is an important variable. Where excellent bonding has been obtained for molded shells, the sphere strength approaches that of a solid graphite structure, indicating that the strength of the fueled spheres is quite dependent upon the mechanical support provided by the matrix. Comparisons of irradiated and unirradiated spheres show little difference for the longer exposures obtained, up to 6×10^{20} neutrons/cm^2 ($E > 0.18$ Mev). Concern still exists that increased shrinkage may be experienced for longer exposures or a harder neutron spectrum. Dimensional and weight data observed in these experiments are shown in Table 8, where dimensional changes of less than 0.1 % and weight changes below 0.01 % probably are not significant. In general, there was little change in the overall dimensions or in weight.

TABLE 8. — Irradiation-Induced Dimensional and Weight Changes in Fueled Sphres.

Capsule	Sphere Designation	Position	Diameter Change [a] (%)			Weight Change (%)
			Pole	Temperate Zone	Equator	
O5Z-6	GA-S10-9	Top static	+0.05	—0.02	+0.07	—0.01
O6X-6	NCF-S2-V	Top static	—0.07	—0.09	—0.31	—0.05
F1-8B-3	SCC-S16-5	1 (top)	—0.09	—0.09	—0.13	—0.03
01-7	NCF-VS14-2	Top static	—0.3	—0.1	—0.2	0.00
05-7	GA-VS11-1	Top static	—0.23	—0.34	—0.23	—0.14
08-7	GA-VS12-12/1	Top static	—0.37	—0.35	+0.04	—0.07
Loop 2, experiment 1	NCF-S4-6	1 (gas inlet)	+0.11	—0.03	—0.76	0.00
Loop 2, experiment 2	GA-VS13-1	1 (gas inlet)	—0.04	—0.06	—0.04	—0.01

[a] Based on average of four readings; changes less than 0.1 % probably not significant in view of limits of precision of measurements.

The release of solid fission products is also of importance for pebble-bed reactors, which obviously cannot provide for a fuel element purge. The graphite powder surrounding the spheres in the capsules, the graphite structure, and the capsule metal surfaces from several assemblies were examined for radioactive products. Although the results are not fully consistent, the principal isotopes identified were Sr, Ba, Cs, Ce, Zr, and Ru. The variations in the relative amounts observed may result from experimental techniques, since no relationship has been established to correlate these observations with the noble gas release or the fuel element design. It is apparent that this important property of the fuel requires additional investigation. It is believed that the techniques for determination of solid fission-product release have now been

improved to the point where satisfactory data can be obtained from current irradiation tests.

3.4.2. — *Thermal Conductivity of Fueled Graphite as Affected by Irradiation.*

Since the irradiation test capsules are instrumented with several thermocouples, including a thermocouple for measurement of the central temperature of the top sphere, it is possible to deduct the overall thermal conductivity of the spheres. Calculations were made for three of the spheres for which the requisite data were available. Short-term, steady-state, spherically symmetrical heat conduction in the sphere and its immediate surroundings was assumed, and the heat generation was calculated. To obtain the heat generation, corrections were made for the modified thermal-neutron flux due to self shielding, the irregular geometry, and the slow variation of the heat generation rate during the test. It was further assumed that the thermal conductivity of the spheroidized powder bed varied from 0.040 to 0.045 w/cm. °C over the temperature range from 800 to 1200° C. The results of these computations are shown in Fig. 8. It is evident that the thermal conductivities vary significantly

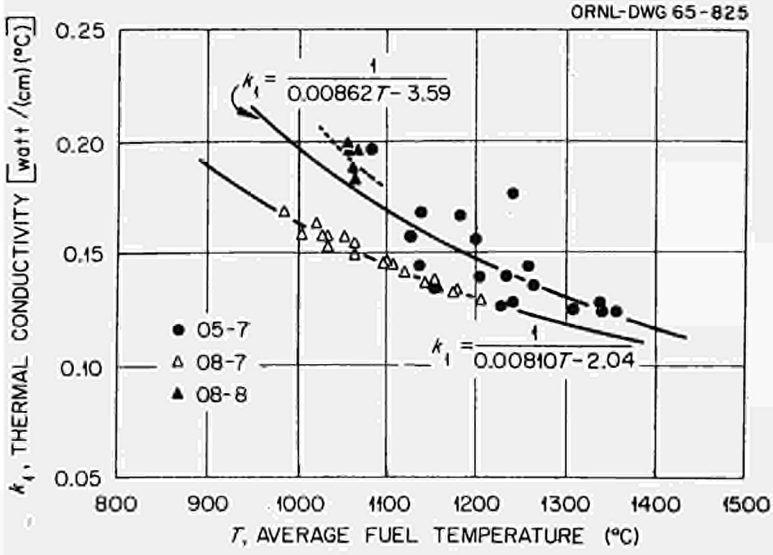

FIG. 8. — Calculated Thermal Conductivity of Irradiated 6-cm-diam, Fueled Graphite Spheres.

with temperature. Little variation with irradiation dose was noted up to 10^{20} neutrons/cm^2 ($E > 0.18$ Mev), except that for one experiment with sphere GA-VS11-2, there was lower conductivity at the end of irradiation than at the beginning. This variation contributes to part of the scatter in the data of Fig. 8. However, this sphere showed relatively poor bonding between the insert and shell, and the observed changes in conductivity seem to indicate a variation in this dimension rather than changes in the conductivity of the matrix.

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FUEL DEVELOPMENT FOR THTR

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ABSTRACT

Several types of spherical graphite fuel elements are under development for THTR. Pyrolytic carbon coated particles serve as fuel.

The different types of elements are :

— A graphite sphere with an annular or spherical grap which is loosely filled with coated particles.
— A hollow sphere which contains a thin layer of coated particles on the inner wall of the sphere and graphite in the remaining hole. The shell consists of electrographite with high strength, especially high drop strength, and good heat conductivity.
— A synthetic fuel ball prepared by heating a moulded mixture of coated particles, graphite powder and binder to 1600° C. A graphite matrix of good properties was developed.

Irradiation experiments in the Risø Reactor in Denmark performed with hollow sphere fuel elements at 2.4 kW and a burn-up to 20 %, showed good stability and low temperature differences between the surface and the centre.

1. — INTRODUCTION.

The fuel elements of the Thorium Hochtemperatur Reaktor (THTR) will be graphite spheres containing uranium-thorium fuel in the form of coated particles.

The extensive THTR programme for development of fabrication methods and irradiation testing of coated particles with oxide and dicarbide kernels and pyrolytic carbon and silicon carbide coatings is not discussed in this report (1), which concentrates on the development of the fuel elements.

The fuel loading for the THTR is not yet fixed. Mainly discussed is a loading of 1 g U^{235} per sphere. The uranium is 93 % enriched and mixed with thorium in a ratio of 10 : 1 for thorium to U^{235} atoms. The fuel element spheres for the prototype reactor AVR which is under construction have a content of 1 g U^{235} and a thorium to U^{235} atomic ratio of 5 : 1. The main barrier for the fission gases will be the coating of the particles. The reactor design calls for fuel elements with a maximum ratio of release rate to birth rate (R/B-value) of 5×10^{-4} for Xe^{133} under reactor conditions. The maximum power per ball will be 3 kW. At a maximum surface temperature of 1050° C the centre temperature of the fuel element sphere shall not exceed 1350° C.

(1) The development of production methods for coated particles at Nukem and the irradiation testing of the particles produced, was very much promoted by the help of the OECD Dragon Project.

For the AVR the maximum power will be 2,4 kW per ball and the corresponding maximum centre temperature 1250° C.

Several types of fuel elements are being developed. They can be divided into two groups : one is characterized by a machined electro-graphite shell of the elements, the other by moulded spheres which are pressed from mixtures of graphite powders, coated particles and binders (synthetic elements). The elements with electro-graphite shell are loosely filled with the fuel particles (elements with annular or spherical gaps) or the particles are fixed on the inside of the electro-graphite shell in a thin, spherical layer (wallpaper type).

All fuel element types provide a fuel free graphite shell surrounding the part which contains the coated particles. In those elements in which the particles are not homogeneously distributed in the centre, the internal core is pure graphite moderator.

The most important goals for the development of the fuel elements are (1) low temperature difference between surface and centre, (2) good strength for thermal stresses and for repeated dropping from several metres height on to the pebble bed, (3) good irradiation stability and (4) low corrosion under reactor conditions.

The heat transfer from the hottest fuel particles near the centre of the sphere to the surface depends on the design of the fuel element and the heat conductivity of the graphite materials. The procedure for selecting electro-graphite materials of good quality, with regard to the goals of development mentioned above, is explained in chapter 3; the corresponding one for moulded graphite materials for the synthetic elements in chapter 4.

2. — Fuel Elements with Loose Particles (in Annular and Spherical Gaps.)

Figure 1 shows the two types of fuel elements with loose particles. The annular gap, which is machined into the electro-graphite sphere, has 25 mm diameter, 25 mm height and about 2 mm width. Compared to other types, this element is relatively easy to produce, even the machining of the graphite sphere with gap and screw. This type is not suitable for normal AVR or THTR fuel elements because the volume of the gap is too small to give space for coated particles with sufficient uranium and thorium (heavy metal).

Table 1 shows figures for tap volume per heavy metal weight for several batches of coated particles. Experiments have shown that the tap density of the particles measured in a cylinder of 2 cm diameter is nearly the same as in a small gap. The tap density cannot be much improved by vibrating or extensive tapping. If the gap is smaller than about 1.5 mm, the ratio of tap density to particle density is lowered from about 0.6 to about 0.5 in a monolayer gap. For THTR, 11 g of heavy metal per ball are required. For melted kernels of 315-480 microns diameter with 120-140 microns coating thickness, about 11 cm^3 gap volume would be required. This yields an annular gap of 25 mm height and diameter, and of 8 mm width. With this width, the heat transfer through the particle layer is insufficient and the central

temperature would become too high. The thermal conductivity of a loose particle layer has been measured in helium up to 700° C. Extrapolation to reactor conditions yields values of 0.005-0.01 cal/cm sec degree centigrade. With 3 kW heat generation

TABLE 1. — Tap volume of several types of melted UC_2 or $(U, Th)C_2$ particles with pyrolytic carbon coatings

Type	Kernel Type	Sieve Fraction (micron)	Coating Thickness (micron)	Content of Heavy Metal (%)	Tap Volume per g Heavy Metal (cm^3/g)
DM1	UC_2	250-500	57	70.4	0.49
DM12	UC_2	300-500	155	44.6	1.29
DM13	UC_2	300-500	117	51.6	0.95
DM15	UC_2	200-300	106	51.4	1.05
DM16	UC_2	160-250	103	41.2	1.58
GM4	UC_2	315-430	100	64.3	0.62
WM4	UC_2	315-430	133	59.5	0.80
WM12	UC_2	315-430	126	56.6	0.68
WM35	$(U, Th)C_2$	315-430	145	43.7	1.26
WM36	$(U, Th)C_2$	315-430	151	51.3	1.07

per sphere, this would lead to a temperature difference across the gap of several hundred degrees C. Therefore the annular gap type element can only be used in special cases when either the gap thickness is reduced to about 2 mm (for instance by using highly enriched uranium without thorium dilution) or the power per ball is low enough to ensure that the maximum temperature does not exceed 1350° C.

Several elements with annular gaps have been tested by irradiation experiments under reactor conditions in the Risö DR3 Reactor. The results are described in chapter 5.

Whereas elements with an annular gap are limited in fuel loading, a new type of graphite sphere with a spherical gap inside is promising. This sphere with spherical

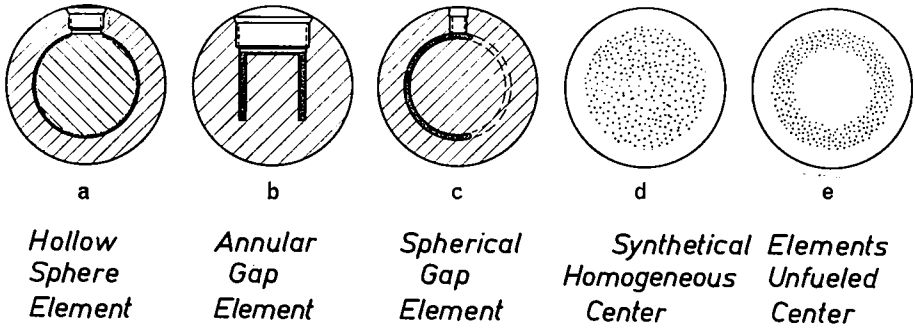


FIG. 1. — Types of Fuel Elements.

gap, proposed and examined by the Ringsdorffwerke, Bad Godesberg, is shown in figure 1 c. The distance between gap and ball surface can be 5-10 mm. The strength of this sphere does not depend upon the shell alone because the shell is supported by bridges to the inner sphere, which divide the gap into several cavities which are all connected to the small filling hole. These gaps are produced by incorporating pieces of volatilizable material during the moulding of the spheres. After moulding, the plastic material is volatilized and the production process finished by carbonising and graphitising the moulded spheres. With a gap of 2 mm width and a shell of 8 mm thickness, about 25 % of the shell can be supported by bridges if 9 cm^3 gap volume is required. With increasing area of bridges, the dropping strength increases.

Both types of the fuel elements with loose particles are difficult to produce in larger quantities if the gap has to be completely filled with particles. This requires very tight tolerances for the gap and for the heavy metal content per specific tap volume of the coated particles. Differences in tap density between different batches of the same production originate mainly from differences in coating thickness, but also from fluctuations of the diameters of the kernels (see Table 1). In a production of this type of element, the dimensions of the graphite spheres, including the width of the gap, will have to be determined before the production of the fuel particles begins. This means that the width of the gap will have to be adjusted to the maximum possible tap volume of the particles.

Variations in the tap density of the particles can be compensated by graphite particles of the same diameter as the coated particles, which are mixed with the coated particles and filled into the spheres. Mixtures of coated particles with graphite powder are not suitable because it is impossible to prevent severe segregation.

3. — Hollow Sphere Fuel Elements.

3.1. — *Graphite Shell.*

Hollow sphere fuel elements have a thin shell of about 10 mm thickness. The thickness of the shell is determined by a compromise between the requirements for heat transfer and strength.

There are several graphite producers in Europe who can provide good graphite types suitable for this type of element. The graphite should have good strength, especially good drop resistance, high thermal conductivity, good irradiation stability and a low corrosion rate in oxidising gases.

The following properties are specified :

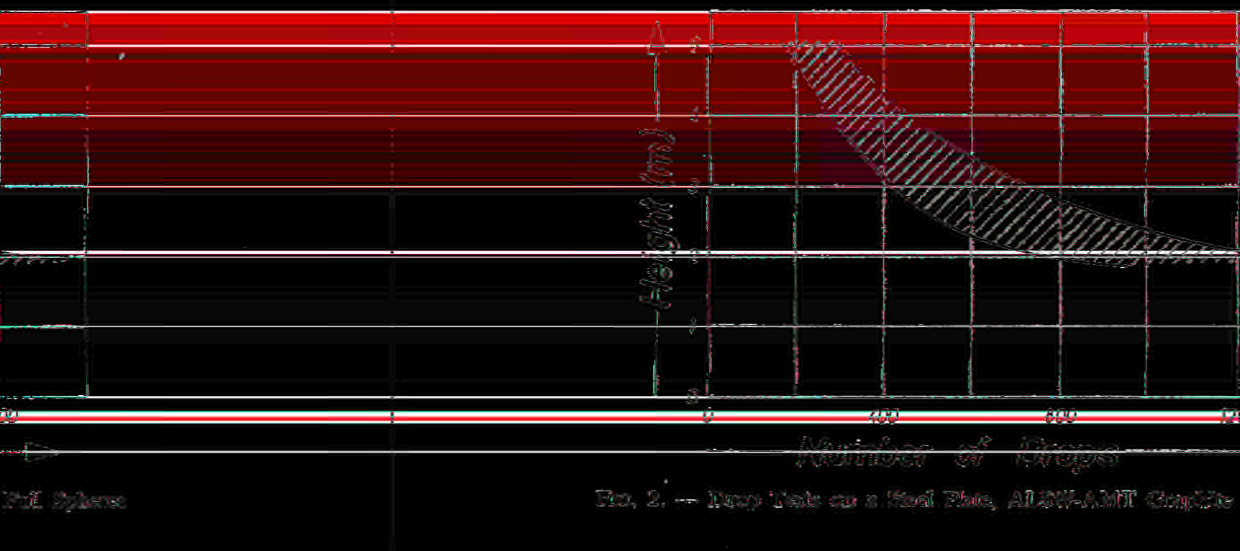
compressive strength	par.	580 kg/cm^2
	perp.	580 kg/cm^2
bending strength	par.	280 kg/cm^2
	perp.	240 kg/cm^2
density		1.70 g/cm^3
heat conductivity at 1000° C		0.08 cal/cm sec° C

The average value for each lot of 3000 spheres shall be higher than the specified

[redacted]

[redacted] ded reactor, through
[redacted] rs and users. There
[redacted] drop spheres, after
[redacted] full sphere show
[redacted] surface. At higher
[redacted] test has been per-
[redacted] ball is charged with
[redacted] to a steel plate are
[redacted] results of drop tests

[redacted] The dropping strength is of high importance for the public
[redacted] this property was until recently unknown to graphite produce
[redacted] be a good difference in dropping behaviour between full and h
[redacted] many repeated drops from normal nature on to a steel plate
[redacted] cracks propagating from the surface and also chipping of the
[redacted] impact tissue the spheres break into two or three parts. The
[redacted] formed in a machine with an automatic elevator. This equipm
[redacted] 16-18 balls simultaneously up to eight drops per minute ar
[redacted] performed with each of these charged balls. Figure 2 shows the
[redacted] from 4-5 m height on to a steel plate.



[redacted] Full Spheres

[redacted] Fig. 2. — Drop Test on a Steel Plate, ALSW-AM1 Graphite

[redacted] 0-70 drops till frac-
[redacted] ture), an extruded
[redacted] moderator spheres

[redacted] Supplementary tests from 12 m on to a steel plate showed s
[redacted] lure. The tests have been made with ALSW/AM1 graphite
[redacted] type of relatively high strength which is used for the extruded
[redacted] in the AVR reactor.

[redacted] [redacted] up to 5, drops from
[redacted] deformed into an
[redacted] ent. In the reactor
[redacted] , where the in pack-
[redacted] the AVR reactor
[redacted] est 50 drops from
[redacted] shows some results
[redacted] and different kind of
(Kingsdorf), which

Hollow spheres with 60 or 12 mm wall thickness failed a
4-5 m on to a steel plate. The impact fissure shows an almost
chipsoidal body and break if the bending strength is not sufficie
fracture does not begin on a steel plate but on a sphere be
is much weaker. Therefore the test drop not specification fo
only requires that 95 % of the fuel elements shall survive a
4 m height on to a bed of spheres without any damage. Table 2
of drop tests with hollow spheres of different shell thickness
diameter. ALSW-AM1 graphite compares with KK 66 graphite

meets the density and strength specifications. The screw diameter is very important for the drop resistance of hollow spheres. The result of impact and compression tests with hollow spheres of ALSW-AMT graphite correlate very well as can be seen in Table 3. Repeated impacts perpendicular to the screw axis produced failing by breaking in two halves through the screw hole, without damage to the screw.

This shows that the bending and tensile strength, measured in the direction normal to the axis of the screw, is the most important value. For this reason moulded graphites are more suitable than extruded ones.

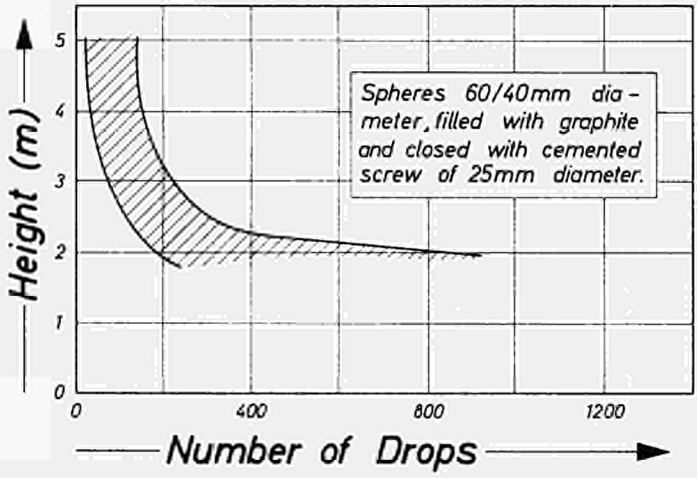


FIG. 3. — Drop Tests on a Sphere Bed, ALSW-AMT Graphite Hollow Spheres.

TABLE 2. — Drop Test with Hollow Sphere Elements.
(Drop from 4 m height on to graphite spheres. The spheres are filled with graphite and closed with a cemented screw.)

	No. of Drops before Failure	No. of Balls Tested
A. *ALSW-AMT graphite spheres*		
1. *Shell thickness 12 mm (sphere 60/36 mm diameter)*		
a) Screw diameter *22* mm	30-210	56
b) Screw diameter *11* mm	127-224	19
2. *Shell thickness 10 mm (sphere 60/40mm diameter)*		
Screw diameter *22* mm	30-130	32
B. *KR 66 graphite spheres*		
Shell thickness 12 mm		
a) Screw diameter *22* mm	177-245	5
b) Screw diameter *11* mm	>700	6

TABLE 3. — Impact and Compressive Strength.
(Hollow spheres ALSW-AMT with 10 mm shell. Impact energy 1 kg from 0.5 m height. Impact and compression direction perpendicular to screw axis.)

Screw diameter	6	10	15	20
Number of impacts before failure	16	9	4	1
Compressive strength (kg)	880	650	570	480

The irradiation stability of the graphite is not so important for spherical fuel elements as it is for long fuel element rods because there is practically no difference in neutron flux in one sphere and the temperature distribution is radial. A small shrinkage of the sphere should be tolerable. Graphite irradiations for the THTR Project are progressing in the BR 2, Mol, in the HFR, Petten and in the Dragon Reactor, Winfrith. The irradiations in Petten and in Winfrith are directed by the Dragon Project in conjunction with Dragon graphite irradiation experiments.

The corrosion rate of graphite spheres in the cooling gas is of great importance because the carbon oxide produced by corrosion may lead to mass transfer and carbon deposition especially in the heat exchanger. The corrosion rates of various graphites are determined in a tube furnace at 1000° C under flowing argon or helium containing one volume per cent of water vapour. The water is added by passing the gas through diluted sulphuric acid. The corrosion rate is measured by determining the difference in weight before and after the experiment and also by measuring the amount of carbon dioxide after absorption in NaOH. The specified maximum value of 70 mg/cm^2, h, atm H_2O can only be reached by very pure electro-graphites with an ash content below 200 ppm.

3.2. — *Fabrication of Hollow Sphere Fuel Elements.*

The coated particles are arranged in a thin layer of about 1-2 mm adjacent to the internal wall of the hollow spheres (wallpaper type, see figure 1). Compared to a homogeneous distribution, this arrangement has the advantage of a shorter distance for heat transfer and hence a lower inpile temperature of the hottest fuel particles.

Fuel elements with homogeneous distribution of particles in the centre of the sphere (injection type elements) are at present produced by UCC as the first loading for the AVR reactor.

The wallpaper type fuel elements are produced by filling a slurry of the coated particles with graphite powder, binder and solvent into the hollow spheres. Subsequently the spheres are tumbled in a special machine in order to produce an even layer of particles. Air is then blown into the sphere while the tumbling continues until enough solvent is evaporated, for the layer to be further dried without tumbling. Proper adjustment of all parameters to the type and amount of particles is necessary in order to get an homogeneous layer.

To study these parameters several types of particles have been used. Particles containing up to 11.5 g of heavy metal can be inserted in one tumbling step.

After complete drying of the layer, the inside of the sphere is filled with natural graphite powder (without any binder) in a special device. With only small pressures of 50-100 kg/cm^2, the density of the filled graphite reaches 1.6-1.9 g/cm^3.

After that, the plug is screwed in and fixed with some binder. Then the binder of the particle layer and of the screw is cracked by subsequently heating the sphere to 1000° C in argon and to 1500° C in vacuum.

Metallographic examinations and leach tests in diluted nitric acid showed that no particles are damaged during fabrication. In metallographic and X-ray tests no small gaps could be found between particle layer and shell. The volume content of particles in the layer is about 60 %. The graphite and binder coke matrix between the particles has a density of about 1.0 g/cm^3. The filler graphite in the sphere centre has sufficient elasticity and compressibility to accommodate to thermal shocks or irradiation shrinkage of the electro-graphite shell. In thermocycling experiments between room temperature and 1200° C no volume change and no cracks have been found.

The irradiation experiments with hollow sphere fuel elements are described in chapter 5.

4. — Synthetic Fuel Elements.

4.1. — *Graphite Development.*

The graphite matrix of the synthetic fuel element differs from electro-graphite mainly because of the lacking final heat treatment. Electro-graphite is heated up to 2700-3000° C. In this process the components of the body, the coke grist and the binder are graphitised. In synthetic fuel elements the heat treatment cannot be raised above 1,700-2,000° C because of the beginning of uranium or thorium diffusion through the coating of the particles. Therefore the graphite body must be formed from a mixture of graphite powder and binder. Since the binder in this material is only carbonised and not graphitised, it is desirable to use as little binder as possible. Natural graphite powder is used as grist together with some artificial graphite powder and sometimes a small percentage of carbon black.

Because of the danger of particle damage during formation at high pressure, the balls are produced by a semi-hydrostatic forming process in a rubber insert die. In this SH process, graphite balls are formed from the prefabricated powder mixture and heat treated afterwards.

The development is not yet completed, but several types of SH graphite produced from different mixtures show very promising properties. The balls have not been damaged in drop tests of up to 1,500 drops on to a pebble bed. The density is above 1.7 g/cm^3, the compressive strength above 400 kg/cm^2, the thermal conductivity at room temperature above 0.15 cal/cm sec °C and the anisotropy very low.

Further examinations and irradiation experiments will be necessary in order to decide which type of element is most promising. Many samples for irradiation between 600 and 1500° C in the Dragon Reactor and in the Petten Reactor have been prepared, together with the electro-graphite samples described in chapter 3.1.

Some years ago graphite samples produced by similar methods were irradiated in the GETR in Vallecitos, USA at 650°. The changes of dimensions and properties have been relatively low.

4.2. — *Fabrication of Synthetic Fuel Elements.*

The synthetic fuel elements have two zones. A shell of 2-8 mm, free from fuel particles, and the centre part with particles. The same graphite matrix is used for the shell and for the centre. First the prefabricated graphite pressing mixture is mixed with the coated fuel particles and then pre-pressed with low pressure to produce the kernel. Afterwards the shell is pressed around the kernel. Both operations are performed in rubber insert dies. The balls obtained have a diameter of 59 ± 1 mm. If tighter tolerances are required, an excess of 1-2 mm is removed by machining.

Fig. 1 shows schematical drawings of two types of synthetic fuel elements : (1) Fuel element with two zones : particle free shell and kernel with homogeneously distributed particles. (2) Fuel element with three zones : particle free shell, particle layer and particle free centre.

The second type provides for better heat transfer and hence for a lower temperature of the hottest particles. This type is produced by filling the rubber die for the pre-pressing of the kernel in two zones. The further fabrication steps are the same for both types.

As mentioned above, the synthetic fuel element needs some further development, especially more irradiation experiments. It will be ready for production in about two years, but then it is expected to be cheaper than the types with machined electrographite shell.

At present, it is difficult to predict whether this will hold true for the costs of the whole fuel cycle of thermal breeder reactors, including reprocessing and refabrication with active fuel.

5. — Irradiation Experiments.

Several fuel elements of the hollow sphere and annular gap type have been irradiated in the DR3 reactor in Risø, Denmark. These tests had three aims :

1. To demonstrate the irradiation stability of the fuel elements.
2. To measure the amount of fission gases released during irradiation in order to find out whether the coated particles have been damaged during fabrication.
3. To show that the centre temperature of hollow sphere elements at a surface temperature of 1000° C and at 2.4 kW power, does not exceed the specified value of 1250° C.

The coated particles for the test elements have been produced by the OECD Dragon project. The sintered (U, Th)C_2 particles had a U^{235} : Th ratio of 1 : 2.7 (92.8 % enriched uranium). They were coated with about 100 microns of pyrolytic carbon. The examinations showed that the coating was of excellent quality.

Each of the three irradiation capsules contained four hollow spheres and three annular gap elements. The hollow spheres have been filled with about 9 g coated particles, containing 0.85 g U^{235} and the annular gap elements with 6.6 g particles with 0.63 g U^{235}.

The irradiation capsules have been supplied by the Dragon project (the proved Fuel Element Replacement Rig), only the internal graphite part had to be redesigned to adjust it for irradiation of balls.

Three capsules have been irradiated — one for one month, one for two and one for three months. The burn-up per irradiation month was about 7 % of the U^{235}.

The hollow sphere fuel element in the region of the capsule with the highest neutron flux yielded 2.4 kW power at a surface temperature of 1050° C and a centre temperature of 1250° C. These were the exact values anticipated in capsule design.

The element with annular gap producing the highest power (near to the centre of the capsule) yielded 1.9 kW with a surface temperature of 970° C and a centre temperature of 1235° C, measured in the centre part of the graphite sphere. As this part has contact to the outer surface of the ball, it does not reach the temperature of the particle layer where the hottest spot has to be calculated about 100° C higher. The centre temperatures of the other fuel elements have been in the range between 960° C and 1250° C, corresponding to flux distribution.

The irradiated capsules were transported to the hot cells in Studsvik, Sweden. The post irradiation examination is not yet completed, but metallography showed that the fuel elements behaved well and none of the Dragon particles had been damaged. Fission gas released from these particles seems to be low, but the figures have to be confirmed.

Further irradiation tests for the THTR irradiation programme are in preparation in the BR2, Mol; the FRJ2, Jülich; the HFR, Petten and the Dragon Reactor, Winfrith.

ACKNOWLEDGMENTS

The extremely helpful promotion of the development of our irradiation tests by the staff of the OECD Dragon Project is greatly appreciated. By supplying coated particles, irradiation capsules and hot cell equipment, the Dragon Project facilitated and accelerated the test programme.

The irradiation tests in Risö have been sponsored by the German Government. BBK and Nukem have been responsible for the programme.

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FUEL FABRICATION TECHNIQUES FOR HIGH TEMPERATURE GAS-COOLED REACTOR FUELS [1]

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ABSTRACT

The fuel for future General Atomic High Temperature Gas-Cooled Reactors will probably be in the form of fission-product-retaining, coated particles. These will be incorporated, either loose or bonded, into a graphite structure which will serve as the fuel element.

The fabrication of these coated particles consists of two steps, the formation of the fuel-bearing seed and the deposition of the coating. Relative merits of « dry » processes and « wet » processes for seed production are discussed. « Dry » processes include agglomeration, followed by sintering or fusion of the agglomerate, and plasma arc processes. The sol gel process is used as an example of a « wet » technique. Of the techniques listed, the agglomeration-sinter or melt combination is the method by which most of the particles to date have been made, while the sol gel method holds the most promise for low cost fabrication.

The seeds are coated by the application of pyrocarbon and/or silicon carbide by pyrolysis of appropriate compounds while the particles are suspended or agitated in some way. This can be done in a fluidized bed, a rotating drum device (similar to a ball mill) or in a vibrating bed. The fluidized bed technique is probably the most developed of the ones listed. Coatings having a great variety of physical and mechanical properties can be deposited by this method by varying the temperature of deposition, the gas flow rate, and the coating gas composition.

There are still a great number of uncertainties in the production processes which can only be resolved by production experience.

The fuel for future General Atomic High Temperature Gas-Cooled Reactors will probably be in the form of fission-product-retaining, coated particles which will be incorporated into long, cylindrical graphite fuel elements. The particles will either be poured (loose) into holes or cavities in the element, or bonded into the graphite by some sort of matrix material such as pitch. As presently envisioned, the fuel elements will be twenty feet long, and about four and a half inches in diameter. The element will be assembled from a series of shorter pieces (about four or five feet long) which will contain the fuel holes (Figure 7). There will be no permeability or back-sweeping requirements on the graphite to be used in the fuel element since the coatings on the particles will be used to retain the fission products.

The particles themselves will contain a seed of either oxide or carbide and will have multiple coatings of pyrocarbon and/or silicon carbide deposited by pyrolysis. The over-all thorium to uranium ratio in the reactor core will be approximately 25 to

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one. Individual particles, however, will not all have the same composition. In order to minimize the uranium-236 content of the uranium-233 to be recycled, it is desirable to separate the thorium and the uranium-235 in the fuel as much as possible. A larger fraction of the particles will thus contain only thorium and a smaller fraction will have a low thorium-to-uranium ratio. The two different types of particles will probably be made in different diameters or have some other distinguishing physical or chemical characteristic built into them to facilitate separation before reprocessing to recover the uranium-233. The nominal diameter of the coated particles will be in the range 250 to 1,000 μ and the coatings will be required to maintain their integrity during reactor core life (several years) in order to retain the fission products.

The Peach Bottom fuel, by contrast, does not require fission product retention in the coated particle. The fission products are kept to an acceptable level in the main coolant stream by backsweeping in the controlled permeability graphite sleeves and the use of a purge gas over the fuel compacts which is led to a separate fission product trapping system. The Peach Bottom core, which has already been delivered to the reactor site in Peach Bottom, Pennsylvania, consists of 804 cylindrical graphite elements, each about ten feet long and about three and a half inches in diameter. More than 2,000 kilograms of uranium and thorium in the form of coated uranium-thorium dicarbide particles were made. These particles have a nominal seed diameter of 200 μ were coated with a single layer of pyrocarbon of about 55 μ thickness, and were incorporated into annular fuel compacts by hot pressing them into a graphite and pitch matrix [1, 2]. These compacts were then placed on a central spine and put into the graphite sleeves which were then fitted with appropriate end pieces. The fuel element is shown in Figure 1.

1. — Coated Particles

The fabrication of fuel elements for future High Temperature Gas-Cooled Reactor will require the fabrication of fission-product-retaining coated particles and their incorporation into a graphite structure. The fabrication of the coated particles consists of two steps : the formation of the fuel-bearing seed, and the deposition of the coating.

The fabrication methods for fuel seeds can be thought of as either "dry" or "wet" processes. The "dry" processes involve the handling of powders or slurries while the "wet" processes involve solutions of colloidal dispersions in a liquid. An example of a dry process is the agglomeration technique using either a V-blender or a gyrating bowl. A fuel-containing compound in the form of a fine powder is put into the mixing container with appropriate vehicles and binding agents and agitated. With the proper amount of vehicle and binder, and the proper agitation speed and time (all determined empirically) the mixture takes the form of small agglomerates having a reasonably narrow size distribution. In the case of the Peach Bottom fuel, oxides of uranium and thorium were blended with graphite powder, trichlorethylene, and ethyl cellulose and tumbled into a V-blender for about two hours. The resulting agglomerates were dried in an oven and screened, the over-

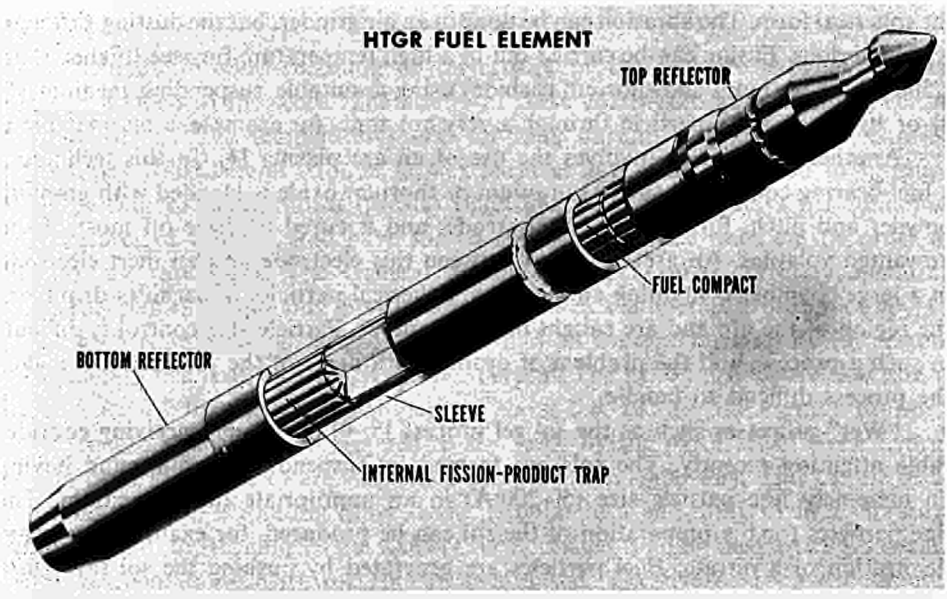

FIG. 1. — Peach Bottom Fuel Element.

and under-sized particles being recycled to the blender. The agglomerates were not particularly round. The operation can also be carried out in a gyrating bowl, in which case the agglomerates seem to have a nicely rounded form which eliminates the necessity for a spheroidizing operation [3]. Agglomerates may also be formed by forcing a thick slurry through a screen of appropriate size. The resulting material may be rounded in a planetary mill, a gyrating bowl, or in an air grinder. There are many variations on the processes listed above, but most of them have about the same advantages and disadvantages. They have the advantage of using relatively simple and inexpensive apparatus, which is an advantage if only small quantities of fuel are required. The processes proceed slowly and can be easily observed; that is, they can be stopped at any time and the product examined without ruining the batch. This slow production rate is, of course, also a disadvantage when one considers the production of larger quantities of fuel. The large amounts of handling and observation would be especially annoying and costly in a remote operation. In addition, the size distribution of the agglomerates is influenced by characteristics of the starting powder, which are difficult to identify and control. The dusting problems require a very special attention because of the toxic nature and value of the materials being processed. The processes can, nonetheless, be made to work and can produce a satisfactory product.

These "dry" processes are generally followed by some sort of a sintering or fusion step. If the agglomerates are rounded, as in the case of the gyrating bowl technique, a simple sintering for densification or conversion to another chemical form (oxide to carbide, for example) is enough to prepare the material for coating. If the agglomerates are not rounded, then they must be either abraded or fused in order to obtain

the spherical form. The abrasion can be done in an air grinder, but the dusting problem is quite serious. Fusion can be carried out in a high temperature furnace (higher than 2,500° C for uranium and thorium carbide) using a suitable suspending medium [1, 2] or by passing the particle through a very hot zone, for example, a plasma torch.

Another dry process involves the use of an arc plasma [4]. In this technique, a fuel-bearing compound such as uranium or thorium oxide is blended with graphite powder and pitch, formed into an electrode, and is cured to drive off most of the unwanted volatiles. An arc is formed between this electrode and an inert electrode in a large chamber containing an inert gas. Spherical particles of carbides drip from the region of the arc and are caught in the chamber. Particle size control is difficult in such a process, and the problem of dealing with dusts of the product also makes the process difficult to handle.

"Wet" processes such as the sol gel process [5, 6] have been receiving considerable attention recently. The sols are formed by suspending a compound having an extremely fine particle size (50-200 Å) in an appropriate liquid medium. The fine particles for the preparation of the sol can be produced, for example, by steam denitration of a nitrate. Fuel particles are generated by pushing the sol through a nozzle. The spherical form of the particle is generated by the surface tension between the sol droplet and the gelling liquid. The sols are gelled by dehydration, changing acidity or by changing the form of the nitrate present. The gel particles are collected, washed, dried and sintered to densify them. "Wet" processes, such as the sol gel process, have many advantages. The process is amenable to continuous operation, that is, it can be accomplished in a series of vessels and pipes that can lead continuously from one to the other without the necessity of intermediate handling. It can thus be controlled by a series of valves, which is especially attractive for remote operation. Toxic materials in the process are handled in solutions or sols, which avoids the dusting problem of the agglomeration techniques. In addition, off-specification materials can be easily recycled by reintroduction into the dissolution step. The gel particles resulting from this operation are very easily sintered to high density (99 % of theoretical) at reasonably low temperatures (about 1,200° C). The particles may in fact be sintered while in contact with one another. The individual spheres densify without sticking to one another. This low temperature densification is a real advantage.

"Wet" processes, as a class, have the disadvantage of using large amounts of fluids, such as wash water. The disposal of these contaminated liquids can be expensive. Sol gel processes, in particular, have the problem of dealing with high surface area materials which can be difficult. The stability of sols is sensitive to small changes in acidity and chemical composition of the suspending liquid which makes control of the sol-forming step difficult. In addition, there is at the present no general recipe for preparing particles with a wide range of uranium-to-thorium ratio. There is also no extensive production experience in the use of this process, which may mean that some of the disadvantages may not yet have been uncovered. More experience and development is necessary, but the "wet" processes seem to possess distinct advantages that will make them the preferred ones in the long run.

2. — Coating

The fission-product-retaining characteristics of the fuel are provided by the coatings applied to the seeds. The ability of these coatings to perform this function satisfactorily at high temperatures and for long burnups has been demonstrated many times. The General Atomic Triplex particles have been irradiated to burnups of 21 percent of metal atoms (greater than 200,000 megawatt days per tonne) at temperatures of 1,400° C and have shown fission gas release rates on the order of 10^{-5} for the rare gases. The General Atomic In-Pile Loop is currently operating with a fuel element containing loose coated particles at a temperature of about 1,500° C. It has been running for 9 months and has had an average burn-up of 1.6 percent heavy metal and a peak burn-up of nearly 3 percent. The fission product release rate is about 5×10^{-6} for the xenon isotopes and about 1×10^{-5} for the krypton isotopes.

Pyrocarbon coatings are applied to the seeds by the pyrolysis of some hydrocarbon while the particles are suspended or agitated in some way. This can be done in a fluidized bed, a rotating drum device (similar to a ball mill), or in a vibrating bed. In each of the techniques the walls of the container holding the particles is heated and a gas containing a hydrocarbon is introduced. The hydrocarbon decomposes on entering the heated zone and the resulting carbon is deposited on the particles. The methods differ in the method used to levitate or agitate the particles. In the fluidized bed device the hydrocarbon gas may be mixed with an inert gas in various proportions and used to levitate the bed of particles. In the rotating drum device the particles are placed in a rotating horizontal drum and are cascaded as in a ball

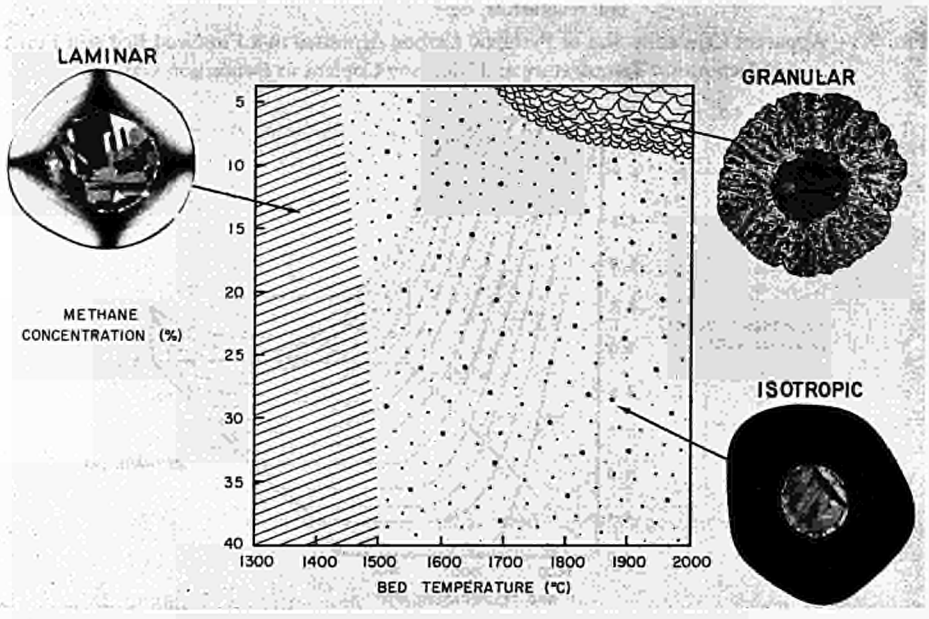

FIG. 2. — Metallographic Appearance of Pyrolytic Carbon Coatings as a Function of Deposition Temperature and Methane Content in Deposition Gas.

mill. The hydrocarbon gas is introduced through along the centerline of the drum. In some experiments done on the vibrating bed, the particles are placed in a vibrating tray. The gas is introduced from the top and impinges on various particles as they come to the top of the vibrating bed.

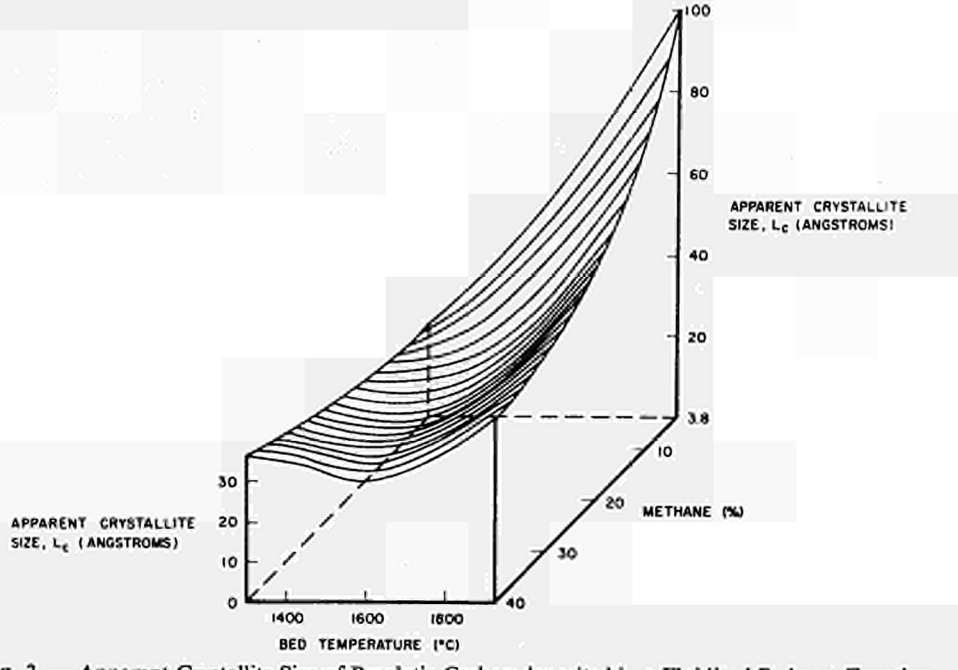

FIG. 3. — Apparent Crystallite Size of Pyrolytic Carbon deposited in a Fluidized Bed as a Function of Deposition Temperature and Methane Content in Deposition Gas.

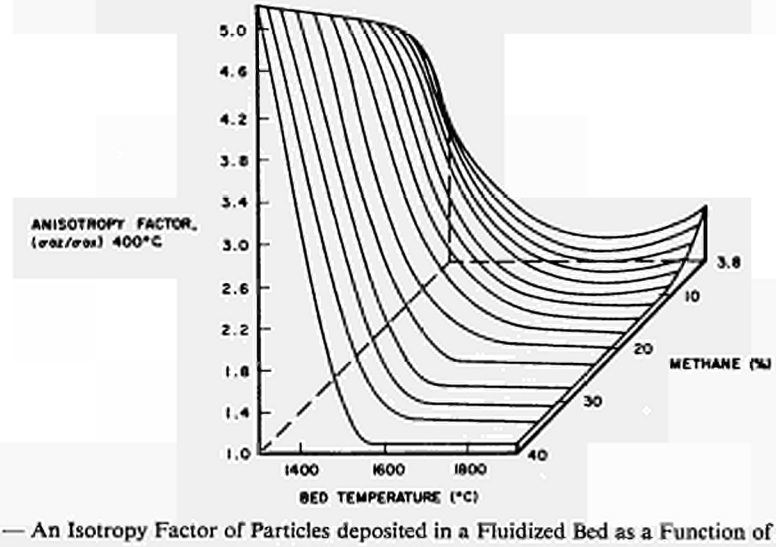

FIG. 4. — An Isotropy Factor of Particles deposited in a Fluidized Bed as a Function of Deposition Temperature and Methane Content in Deposition Gas.

There has been an extensive amount of work done on the properties and characteristics of the pyrocarbon deposited in a fluidized bed [7, 8]. By varying the temratures of deposition it is possible to deposit highly oriented pyrocarbon with varying degrees of preferred orientation, crystallite size, flexure strength, modulus of elasticity, density and metallographic appearance. This is illustrated in Figures 2 to 6. It is evident from these figures that a wide degree of control is available. In addition,

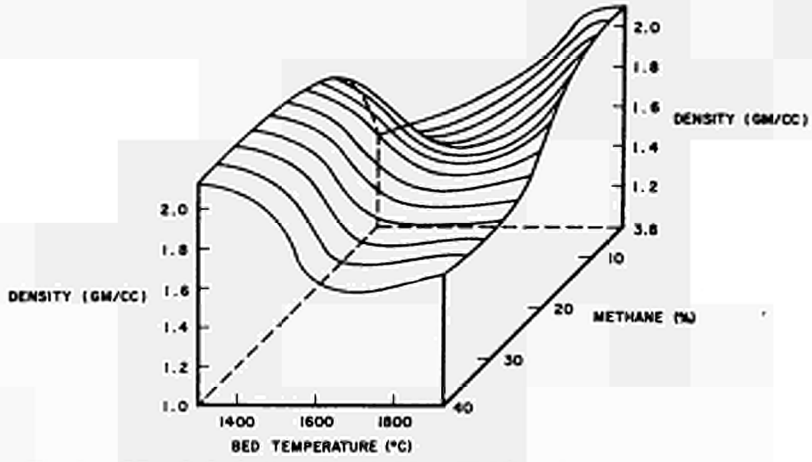

FIG. 5. — Density of Pyrolytic Carbon deposited in a Fluidized Bed as a Function of Deposition Temperature and Methane Content in Deposition Gas.

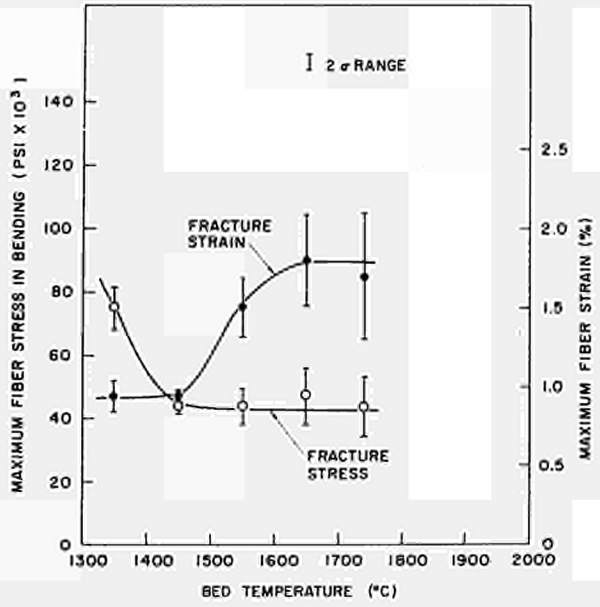

FIG. 6. — Maximum Fiber Stress and Strain at Fracture for Pyrolytic Carbon deposited in a Fluidized Bed as a Function of Deposition Temperature at a Methane Concentration of 40 % in Helium Carrier Gas (Total pressure one Atm).

it has been found by experience that there is very good uniformity of coating thickness among the particles in a batch and that the coating process can be controlled to give a desired thickness within a few microns. The process scales up nicely and except for criticality limitations, there is no reason why a very large coater couldn't be built to handle tens of kilograms at a time. At this time coaters have been operated up to five inches in diameter and have coated seven kilograms of particles in a batch. In future production it will be necessary to separate thorium-containing particles from uranium-containing particles, so that the criticality limitations will be removed from most of the material manufactured for a core. Very large fluidized bed coaters are thus in the realm of possibility. On of the disavantages of the fluidized bed coater is, of course, that it uses large quantities of inert gas which has to be disposed of or regenerated in some apparatus and recirculated. In either case there is an added expense. Rotating drum coaters have the advantage of needing no addition of inert gas for levitation of the particles. They are, however, more complex mechanically and might present difficulties in remote operation. At the present time the status of knowledge and the degree of control available in the fluidized bed coaters seems to favor them over the rotating drum coaters for the deposition of pyrocarbon coatings on fuel particles. There is very little known at the present on the operating characteristics of vibrating bed coaters.

3. — Fuel Element Assembly

Incorporation of the particles produced by the methods indicated above into the graphite fuel element should be a relatively simple affair if they can be poured in and used as a bed of loose, coated particles. The particles would be poured into holes drilled into a section of graphite and provided with some form of closure. The short sections of fuel element would then be joined together to form a larger, long fuel element. Alternatively, the particles could be bonded into the element in some way. In this latter case, this could be done by the use of pitch or other carbonaceous resin. Ground pitch can be mixed with the particles as they are poured into the element. This pitch would be carbonized in a subsequent heating operation and would act as the binder. Alternatively, the individual coated particles could be given a coating of pitch in a simple spray coating device and then poured into the element. The particles would then once again bonded by heating the resulting bed of particles.

4. — Costs

It is very difficult indeed to predict the cost of a High Temperature Gas-Cooled Reactor fuel element with precision. The use of fission-product-retaining, coated particles has removed the need for control of the permeability of the graphite and removes some of the uncertainty in that area. There are, however, a great number of variables in the particle production processes which have a marked influence on the cost. The influence of these variables can only be determined by extensive experience in production. As an example, in the production of seeds, the fraction of yield in the

desired size range and the experience on material loss has a very large effect on the cost of the particles. This is especially true in the case of the particles containing fully enriched uranium and the amount and nature of the material to be recycled. These losses can only be determined during a large, production run. It is reasonable, and even necessary, to make estimates of the costs, but I would caution against placing great reliance on these figures until a substantial quantity of fuel has been produced.

We feel that fuel for a 1,000 megawatt (electrical) capacity General Atomic High Temperature Gas-Cooled Reactor can be fabricated in the next few years for about 200 dollars per kilogram (of heavy metal) for the particles plus about 500 dollars per element for graphite and assembly. Oak Ridge National Laboratory in a recent report has shown calculated fabrication costs considerably lower than these figures. These costs are calculated assuming a plant that supports 15,000 Mw(e) of generating capacity and also assumes a reasonably well established production scheme, that is, one which has been scaled up and is running smoothly. We feel that the costs that they have given are achievable and that, in time, they will be achieved. Accurate prediction of costs, however, for fuel to be fabricated during the learning period in fabrication technology is hazardous.

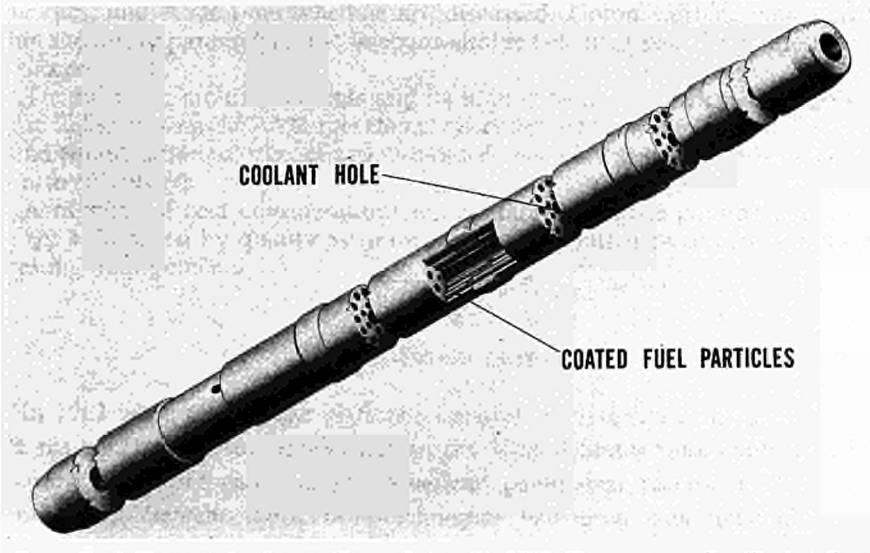

FIG. 7. — Fuel Element for Large General Atomic High Temperature Gas-Cooled Reactors.

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DESIGN AND FABRICATION OF THE ARBEITSGEMEINSCHAFT VERSUCHS-REAKTOR (AVR) INJECTION-MOLDED FUEL ELEMENT

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ABSTRACT

In 1962 when Oak Ridge National Laboratory first turned its attention to the AVR reactor, several fuel fabricators in the United States were contacted. During earlier work on the American pebble-bed reactor concept, a number of fuel element fabrication techniques had been experimentally explored by these companies. This experience could be immediately transcribed to the AVR fuel. Various fuel element design concepts were available for consideration, all embodying coated particles which had already demonstrated satisfactory fission product retention at temperatures and fluxes anticipated in the AVR reactor.

A description of the evaluation of the various designs in terms of unique AVR requirements, previous reactor tests on materials to be used, present state-of-the-art, economics, and AVR time schedule are discussed. Union Carbide concluded that, within the above parameters, the injection-molded element had the greatest potential for success.

The injection-molding process and its unique features which make it so suitable for use in fabricating the AVR fuel elements are described in some detail. Advantages, of one modification of the injection-molded element, the unfueled-center design, are also discussed.

A number of cost considerations in fuel fabrication are pointed out, emphasizing the role payed by quality assurance provisions, either in design, specifications, or testing requirements.

1. — INTRODUCTION

In 1962 when Oak Ridge National Laboratory first turned its attention to the AVR reactor, several fuel fabricators in the United States were contacted. During the course of earlier work on the American pebble-bed reactor concept, several potential fuel element fabrication techniques had been experimentally explored by these companies. This experience could be immediately transcribed to the AVR fuel. Among the designs which had been explored were molded admixed spheres with and without an unfueled co-molded shell, machined shells mechanically enclosing an admixed core, and machined spheres containing loose particles. Needless to say, all designs embodied coated particles which had already been tested satisfactorily at temperatures and fluxes anticipated in the AVR reactor.

The dominating properties required of the fuel elements were abrasion resistance, steam oxidation resistance, and impact resistance, with irradiation stability, thermal

conductivity and cost as important side considerations. An evaluation of the various designs and fabrication techniques at an early period in the AVR program appeared in our view to be as follows :

TABLE 1. — Ranking of Element Concepts.

Element Type	Cost	Abrasion Resist.	Impact Resist.	Steam Oxidation Resist.	Irradiation Stability	Heat Transfer
Admixed Sphere Without Shell	1	4	4	4	3	1
Admixed Core, Co-Molded Shell	3	1	3	1	4	3
Machined Sphere, Loose Particles	2	2	1	3	1	4
Machined Shell, Molded Core	4	2	1	2	1	2

In 1963 Union Carbide formed its Nuclear Products Department around the Carbon Product Division research and development personnel who had been exploring fuel element fabrication and had successfully developed the Duplex coated particle. This augmented group decided to re-evaluate the various sphere designs and seek a less expensive fabrication technique which would take advantage of as many known factors as possible. It is the purpose of this paper to review these design criteria and describe the resulting fabrication techniques currently being used to manufacture the AVR element.

2. — DESIGN EVALUATION

In reviewing the state of the art in early 1963 as indicated in the above table, we quickly reached two conclusions. First, the admixed sphere without shell could be immediately rejected for the AVR application. The occurrence of fuel particles in the immediate vicinity of the sphere surface, with or without leaching, could not be tolerated on the basis of the required abrasion, oxidation, and impact resistance.

The second immediate conclusion rests on a slightly more obtuse reasoning. One could add two more columns to the above table, namely, the time to reduce a development concept to a practical production item and the time to demonstrate by reactor tests the necessary performance quality. The procedure for molding an unfueled outer shell around a fueled admixed core had been demonstrated in the laboratory. There seemed little question that such a technique could be carried into production although economics were open to some question. However, in view of the AVR schedule, the time required to prove radiation stability with regard to strength, dimensional changes, and thermal conductance precluded this as a feasible approach.

We were left with two design concepts to consider as a practical route in the time available. Both were based on a machined sphere, and both had strong advantages. The outer shell could be machined from a graphite of well-known physical properties, and thus offered no apparent problem in terms of radiation stability, thermal conductivity, or basic mechanical properties. As a potential candidate material among those manufactured by Union Carbide, we selected grade ATJ. This is an extremely fine-grained essentially flaw-free graphite with well-known properties. Impact tests at Oak Ridge [1] and in our own laboratories showed that this material was satisfactory. Steam oxidation tests indicated this grade was extremely marginal. However, if necessary, purification should eliminate this problem. On these bases we elected to use grade ATJ as the shell material for all subsequent machined-shell development.

Fragmentary irradiation data available from unpublished Hanford experiments indicated that grade ATJ behaved comparably to the "good" nuclear graphites of the AGOT class, exhibiting perhaps a 50 % greater contraction in the intermediate temperature range [2]. On the basis of raw materials and fabrication techniques, we believed the radiation stability would persist to higher temperatures. Subsequent experiments by Oak Ridge National Laboratory have verified this conclusion [3]. At a neutron exposure of 1.5×10^{21} nvt (> 0.18 MeV) in the 900-1,000° C temperature range, the liner contraction of this graphite was approximately 1 %, corresponding to 0.6 mm for a 6-cm diameter AVR sphere. This is well within the reactor requirements.

Because of the sharp dependence of fission product release on temperature for coated particles, a loose particle design opened up numerous questions. Although the thermal conductance of packed coated particles was not known at the time, we feared it would be so low as to force a reduction in fuel loading per ball in order to safely limit the individual particle temperatures. This in turn would cause a change in the total number of elements per loading and require a complete re-evaluation of the core calculations. Therefore, we felt constrained to search for a solution based on an admixed-core machined-shell design.

The original machined shell concept, which was eventually carried to an advanced and refined design by Oak Ridge National Laboratory, consisted of a molded fuel insert which was contained in a split graphite shell consisting of essentially two hemispheres. A number of problems existed with this basic design:

a. Impact Resistance : A split shell (two hemispheres) joined by threaded, bayonet, slip, or other mechanical means presents a large machined-surface-to-machined-surface contact and sharp angles. This willl necessarily result in an excessive number of notches with localized stress upon impact. In addition a good mechanical contact between insert and shell for distributing stress upon impact is difficult to obtain. This requires that either the internal cavity of the shell or outside dimensions of the insert be fabricated to extremely close tolerances, both costly, or that a buffer layer connecting shell and insert must be provided.

b. Heat Transfer : Good physical contact is also a necessity to achieve good heat transfer from the fueled insert to the shell. Cost considerations regarding shell and insert tolerances are the same. The use of a graphite cement buffer might provide this contact; however, a cement consists of an extremely high binder content which would only be carbonized, leaving a porous amorphous low thermal conductivity layer. In the presence of high temperature neutron irradiation, this layer would probably undergo considerable shrinkage resulting in a gap.

c. Insert : A large number and variety of fueled matrix materials had been developed and tested. Although molding techniques had been perfected by which spheres could be fabricated to close tolerances without excessive flashing, it appeared that achieving good contact with the shell would still require machining the insert. This would necessitate a subsequent costly leaching to removing exposed uranium and thorium.

From an examination of the above difficulties and a review of the excessive number of operations required in the fabrication, we concluded that not only would the split-shell element prove to be marginal, but would also be excessively costly.

At this point, in February, 1963, our opinions had crystallized. We desired an ATJ shell with an improved geometrical design which would eliminate machining problems as they pertained to impact resistance, a fueled insert which would avoid the dimensional matching problem, and an overall process which would be considerably less expensive than the existing concepts. This was the problem presented to our technical staff, and a company-sponsored development program was immediately started.

3. — The injection-molding Process

The obvious method of minimizing the impact resistance and notch sensitivity problems was to machine a complete hollow sphere with as small a hole as possible, which could be later plugged. The machining operation could be simplified for a 1-cm walled sphere if the hole were kept to a minimum diameter of approximately 2.25 cm. The machining could be accomplished with a fly-cutting tool, contoured to the desired inner radius of the sphere. The tool is inserted into a hole drilled in a solid sphere and then displaced to hollow out the interior. A single-lead modified British Whitworth thread was selected, again to minimize notch sensitivity.

ATJ spheres of this design could be machined for one-half to two-thirds the cost of the split "hemispherical" design. The resulting basic configuration is shown in Figure 1.

The core-loading techniques for such a shell configuration were not difficult to visualize. The technique was essentially borrowed from the plastics industry, and is frequently termed "injection-molding". In this process, the internal spherical cavity of the shell becomes the mold. The steps used in the process can be described as follows :

a. The ATJ shell is inserted into a support containing two hemispherical cavities of the same dimension as the outer diameter of the shell.

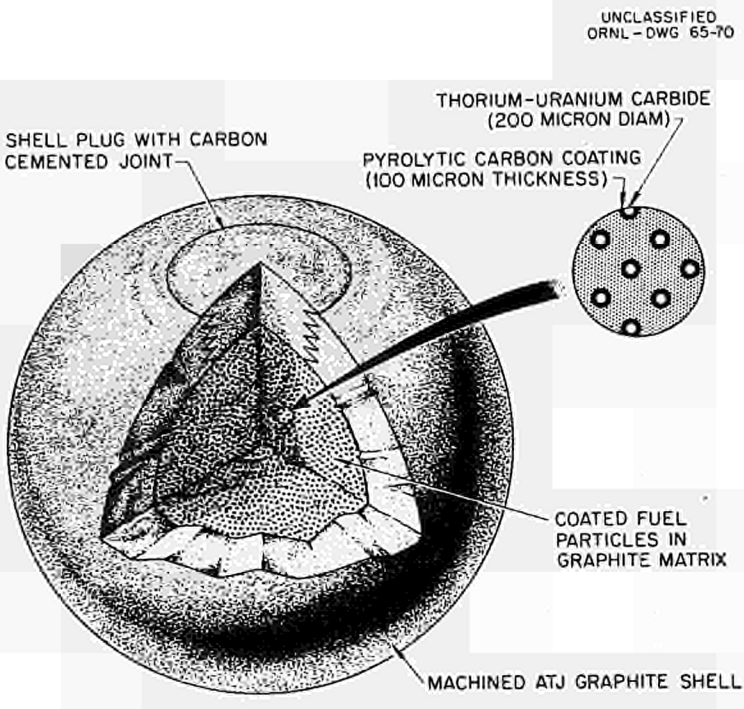

FIG. 1. — Configuration of the Injection-Molded AVR Fuel Element.

b. A tube is inserted into the hole in the shell. It is loosely filled with a carefully weighed-out, premixed blend of coated fuel particles, graphite filler, and thermosetting binder.

c. A plunger contoured to complete the inner radius of the core forces the blend into the hollow sphere. The depth of plunger movement is closely controlled to complete the form of a spherical core. With this type tooling, elements can be filled at the rate of about one per minute.

d. The filled sphere is removed from the tooling, and a threaded, cement-coated plug is screwed into the sphere hole.

e. The sphere is cured to thermally set both the core and the plug cement.

f. The excess portion of the plug is removed by a contoured cutter conforming to the outer radius of the sphere.

g. The sphere is baked to a minimum 1,450° C to establish a carbonaceous bond and essentially eliminate any residual hydrogen.

The injection-molded element is obviously less costly to manufacture from several standpoints. First, as mentioned above, the shell can be machined for about one-half to two-thirds the cost of a split shell. Not only have the tolerances been eased, but the number of fabrication operations required has been significantly reduced; hence the number of inspection steps is fewer.

This design also overcomes some of the inherent disadvantages of a split shell design. An excellent mechanical and chemical bond between the shell and matrix can be maintained. This enhances impact resistance to a considerable extent in addition to promoting good thermal conductivity from matrix to shell.

The above considerations were meaningless unless a suitable fueled matrix could be devised. Normally, a formed graphite body would shrink, thus producing a gap between the fueled core and unfueled shell. Quite aside from the effects on heat transfer, our experiments with impact resistance of hollow balls indicated a gap could not be tolerated.

The solution to this problem was proposed by Mr. A. A. Kellar, and brought to practical fruition by Mr. Kellar, Dr. J. T. Meers, and Mr. R. A. Howard. The solution was both ingenious and effective.

The exfoliation properties of graphitic compounds has been known scientifically for years [4]. A similar phenomenon, also well-established, exists in the graphitic residue compounds. This exfoliation represents a net volume expansion which can be well controlled for the residue compound. Milled purified natural graphite was reacted to the bisulfate and converted to the residue compound. By a judicious admixture of the residue compound with normal powders, it is possible to produce matrix structures which will contract, remain dimensionnally stable, or expand as they undergo subsequent thermal treatment. Representative curves of the thermal

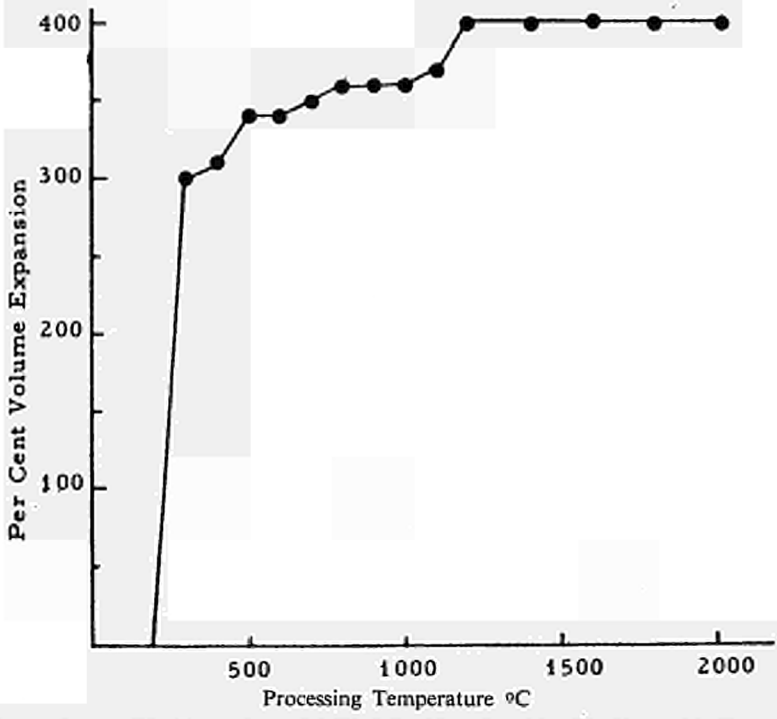

FIG. 2. — Irreversible Expansion of Milled Residue Graphite Bisulfate with Temperature.

behavior for the residue compound and a practical mixture of residue compound and normal graphite filler are shown in Figures 2 and 3.

From the known radiation characteristics of natural graphite, there was no reason to anticipate any problem with these matrices. Irradiation results on the fabricated fuel element have established that this type of matrix is well-behaved

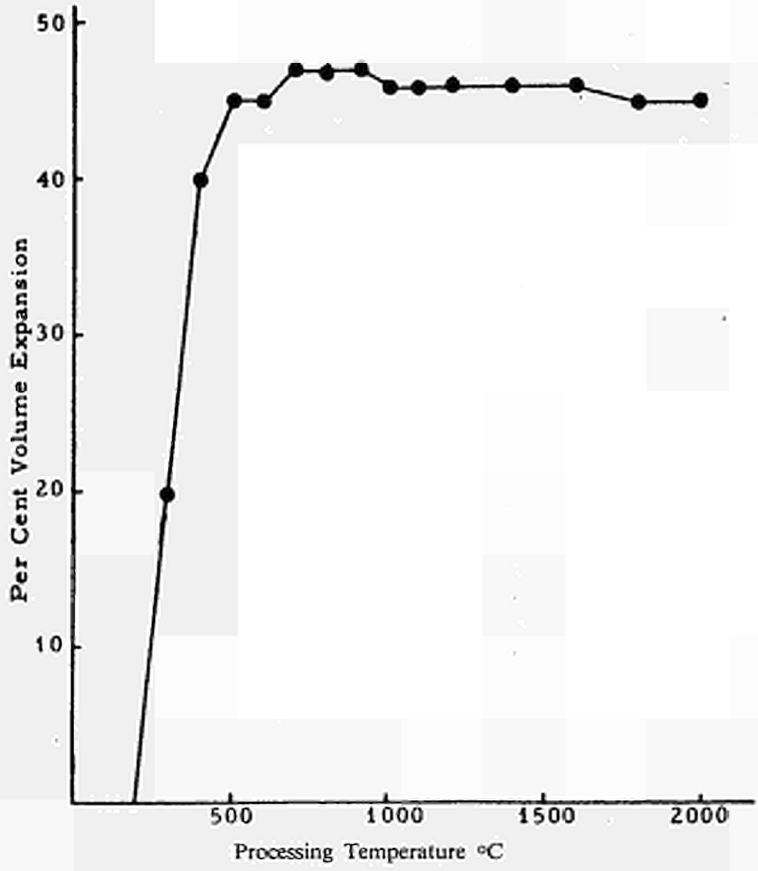

FIG. 3. — Irreversible Expansion of Graphite Matrix Formed from Blended Normal and Residue-Compound Flours.

TABLE 2. — Fuel Matrix Properties - Injection - Molded Elements.

Density	1.55 g/cc
Sonic Modulus	0.700×10^6 lbs./in.^{2}
Crushing Strength	4,000 lbs./in.^{2}
Coefficient of Thermal Expansion (20-300° C)	6.4×10^{-6}/° C
Thermal Conductivity	$.075 \frac{\text{gm cal-cm}}{\text{cm}^2 - {}^\circ\text{C} - \text{sec}}$

and that no gap develops with time as the core matrix undergoes radiation damage. Temperature measurements indicate also that there is no abnormal change in the thermal conductance of these matrices with irradiation.

In Table 2, order-of-magnitude properties of the expanding matrix are given.

4. — The Unfueled-center Design

There are obvious advantages to concentrating the heat sources of a spherical fuel element as closely to the sphere surface as possible. Such a geometric rearrangement of the fuel in the present instance must be consistent with restrictions of local power dissipation and freedom from surface contamination. Suggestions in this

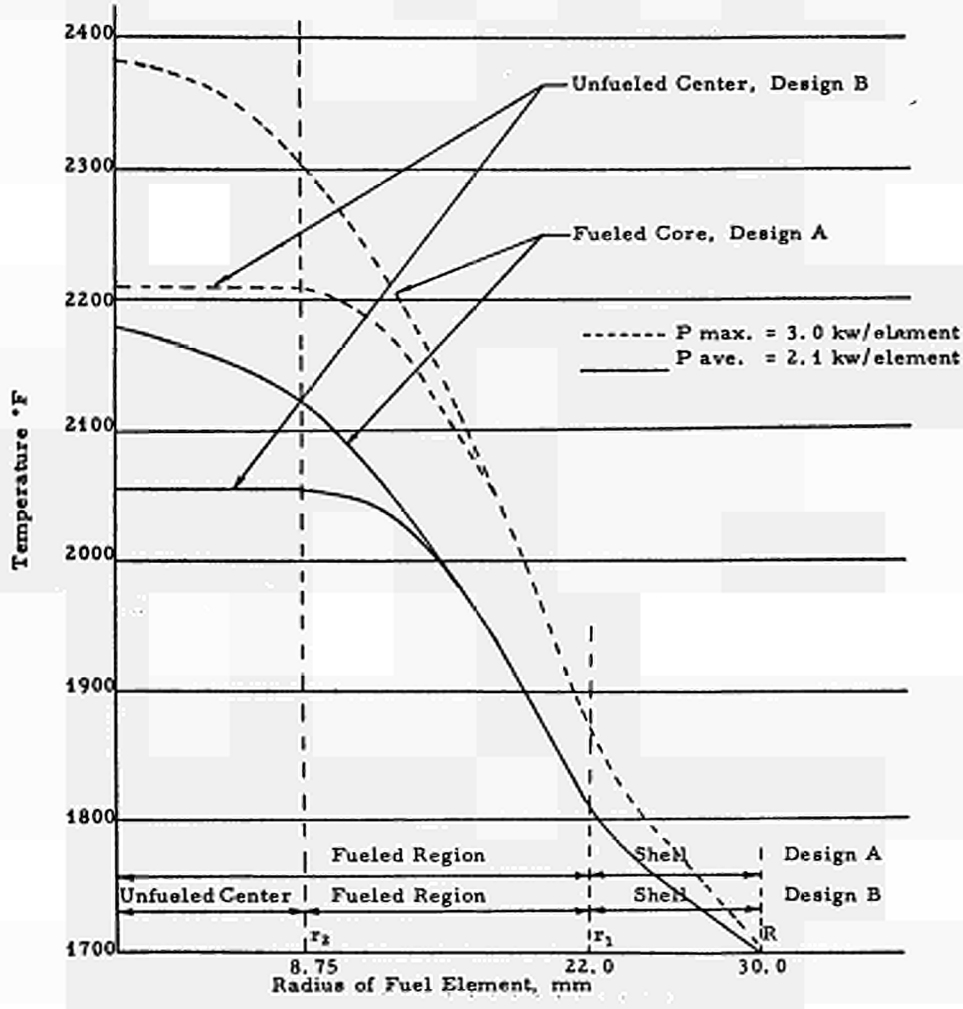

Fig. 4. — Temperature Distribution in Spherical AVR Fuel Element.

direction have originated from numerous sources, of which we are aware of at least the following independent proposals : Brown-Boveri/Krupp, Oak Ridge National Laboratory, Kropfmuhl-Ringsdorff, and ourselves.

Theoretical calculations based on known material properties indicate significant advantages can be gained for an unfueled-center element design. Figure 4 gives the temperature profiles for a solid fueled core (Design A) and an annular fueled core (Design B). Calculations were based on two assumed power levels, 2.1 kW per ball (solid lines) and 3.0 kW per ball (dotted lines). Two conclusions can be drawn from these curves : First, at the presently rated power level of 2.1 kW per ball, the maximum central temperature can be reduced from approximately 2,200° F to 2,050° F with a concomitant increase in the fission-product retention capability of the coated fuel particles. Second, comparing the fueled center at 2.1 kW with the unfueled center at 3.0 kW, it is seen that the same central temperatures can be maintained but with perhaps a 40 % increase in power density.

A number of such elements have been made to demonstrate their production feasibility. The technique is similar to that described in Section 3, with the addition of a further step in the molding procedure. Approximately three-fourths of the core material is injected into the hollow sphere after which a tool introduces a cavity in the injected material. A small solid ATJ graphite sphere is dropped into the cavity, and the remainder of the core material injected. After a few trials it was found possible to locate the unfueled core within 0.010 inch of the true ball center.

The cost of the annular-type fuel element fabricated in this way would add approximately 2½ % to the cost of the all-fueled core element.

5. — Some Cost Considerations

As a strictly commercial fuel fabricator it is not our intent, for a number of obvious reasons, to discuss our experience with specific fabrication costs. However, we can emphasize several cost factors which we feel played a part in the cost of the AVR fuel and which will be vital to fuel fabrication costs not only in future AVR-type loadings, but other reactors.

By careful and extensive development and design, one can make significant reductions in fabrication cost. This we can measure by our price estimates of the split-shell design versus the injection-molded design. Our estimate of the split-shell fabrication was approximately $40.00 per element. The actual price of the injection-molded sphere is $23.00 per element. The difference is largely due to the simplification of fabrication steps, as already pointed out.

However, there remain significant potential savings that can be achieved without altering the basic injection-molded design. Almost onehalf the cost of the present AVR elements is due to the quality assurance requirements. This in no way is meant to imply that the quality and control provisions for the first AVR fueling are too conservative. It is recognized that this fuel represents a "first time" and that this "forerunner" reactor is experimental. Overcautiousness is an initial necessity.

Quality assurance, as we view it, consists of three factors — the technical requirements or specifications placed upon the product, the magnitude of testing and inspection required of the vendor, and the control exerted by the buyer upon the fabricator's process, records, and inspection. Each factor to varying degrees is necessary for adequate assurance of quality; however, their contribution to the overall cost of fabrication is all too frequently overlooked. It is suggested that reactor designers, either in developing fuel specifications and procurement documents or in making advanced design cost studies, recognize the full cost impact of these quality assurance provisions.

It is possible that the savings resulting from unique and ingenious fuel element design, from both performance and economical fabrication standpoints, can be more than offset by stringent specifications, test requirements (particularly destructive), requirements for retention of samples for reference, or information reporting. A greater understanding of materials and which requirements are absolutely necessary is helpful. The same attention paid to designing a fuel element must also be utilized in designing specifications for procurement.

We would also like to recognize a number of other factors which will influence reductions in fabrication costs for future AVR-type fuels. Due to the particular nature of the AVR reactor and the time, state-of-art relationship in the case of the AVR, compromises between fuel fabrication and reactor design were not possible.

a. A fuel fabrication plant designed specifically for AVR-type fuel elements would lead to lower capital changes and more efficient use of space and personnel. This is particularly true if more reactors utilizing an identical fuel element are constructed.

b. The spreading of fuel element procurement over a long period of time so as to provide a continuous production load will also significantly reduce costs.

c. Design improvements, such as the unfueled center, will increase fuel performance at negligible increase in costs.

d. Improvements in coated-particle processing have been continuous over the past few years. In particular, promising experiments with uranium oxide spheroids offer a less expensive starting material than uranium dicarbide.

e. Although the other fuel designs discussed above were dropped from our development program for the first AVR loading, some of these design concepts offer cost savings if specifications are changed. Of particular interest in this regard is the admixed core without a shell and the loose particle types (the last because of their anticipated reprocessing advantages).

We would conclude this paper by stating our confidence in the future of the high-temperature gas-cooled reactor. We are convinced that the fuel fabrication industry is far from achieving the optimum in fuel economics today for this reactor system. The same strides made over the past few years are certain to continue or be surpassed during the next few years.

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SESSION I :

FABRICATION METHODS AND COSTS OF FUEL ELEMENTS

Introduction prepared by D. TYTGAT, Euratom, Brussels

I. — Introduction

As indicated in the program, this presentation is based on data presented in 5 papers from Dragon, Oak Ridge National Laboratory (ORNL), General Atomic, Nukem and Union Carbide.

The original concept for a fuel element of a high temperature gas-cooled reactor was based on "complete release of fission product poisons, but experiment and experience showed that this was impracticable".

In the same time, the development of fission product retaining fuel by using coated particles has introduced a radical change in the fuel concept. "The fission product retaining fuel and fuel elements consist of a series of barrier and sinks designed to prevent or delay the escape of fission products into the primary circuit. The most important barrier is achieved on a microscopic scale by coating of individual fuel particles (250-500 microns diameter)".

While using both coated particles, two concepts can be used for the fuel elements :

(1) the purged fuel element where the release of gaseous fission products in the primary circuit is avoided by using a He purged flow inside each graphite sleeve [Dragon, General Atomic (G.A.)],

(2) the unpurged fuel element.

While some reactor concepts, like the ones using pebbles, cannot use the purged fuel element, the others have the choice between purged and unpurged. Recent trends indicate a preference for unpurged fuel elements. Dragon thinks even to use the "direct cooling" of the fuel inserts, avoiding the use of a graphite sleeve and passing the helium flow directly on the fuel inserts prepared by overcoating the coated particles.

II. — Fabrication routes for coated particles

Coated particles are obtained by coating a kernel of fuel materials prepared by different methods.

A. The fabrication routes for kernels

The fabrication routes may be listed as follows :

(1) Powder metallurgy processes;

(2) Massive carbide routes;

(3) Sol-gel processes.

1. — *The powder metallurgy processes.*

1.1. — Carbide preparation.

Basically these processes use a mixture of Th and/or U fine oxide powders with carbon powder and a suitable binder and have been described in papers published before this meeting.

Different techniques are used for making primary particles, for instance blenders of different types, gyrating bowls, mixing-extruding-chopping technique.

After drying, screening permits selection of the suitable range of particles desired, the oversized and undersized particles being recycled. To obtain spheroidisation of the particles, it is possible to melt the particles, using for instance :

(*a*) A plasma jet (at 15,000° C);

(*b*) A "bed" of carbon powder heated by induction (at 2,500-2,550° C).

If sintering of the kernels with controlled porosity is preferred to melting, spheroidisation must be obtained before any heat treatment. This spheriodisation is necessary, as indicated by ORNL, for spheroidal particles generally perform better under irradiation than irregularly shaped particles with sharp edges and protusions.

The powder metallurgy processes have been the most used up to now by the different laboratories and industries. As General Atomic states "there are many variations on the processes listed, but most of them have about the same advantages and disadvantages. They have the advantage of using relatively simple and inexpensive apparatus, which is an advantage if only small quantities of fuel are required. The processes proceed slowly and can be easily observed, that is, they can be stopped at any time and the product examined without ruining the batch. This slow production rate is, of course, a disadvantage when considering the production of larger quantities of fuel. The large amount of handling and observation would be especially annoying and costly in a remote operation. In addition, the size distribution of the agglomerates is influenced by characteristics of the starting powder which are difficult to identify and control. The dusting problems require a very special attention because of the toxic nature and value of the materials to be processed. The processes can, nonetheless, be made to work and can produce a satisfactory product."

1.2. — Oxide preparation.

The oxide particles studied for the HTGR fuels have been mainly produced by the same agglomeration techniques and by sol-gel techniques which are discussed hereunder.

2. — *The massive carbide routes.*

These routes are described only by Dragon, but are discarded today for the reasons explained in the paper.

3. — *The sol-gel processes.*

The sol-gel technique is very useful in so far as it leads to particles which are very spherical in shape in a much simpler way than by the powder metallurgy route, as illustrated in Fig. 1 of the Dragon paper. It is particularly appropriate for remote fabrication of thorium fuel containing U^{232} (this will be discussed in Session II).

As we said before, the main accent for primary fabrication by the different laboratories and industries has been on dry methods and outside Oak Ridge, limited experience on sol-gel is available at Kema, Dragon or Nukem. One question which might be interesting to discuss is to what extent sol-gel might be more economical than dry methods for a primary fabrication. Should its development be accelerated and adopted for any large primary fabrication plant due to its interest in refabrication? Or could it be reasonable to suppose that any reactor fuel fabrication would need the two methods working in parallel?

Some further questions can be raised :

(*a*) Can the particle size chosen for a high temperature reactor fuel affect the choice between dry or wet methods? Dragon thinks that the sol-gel route seems at the present confined to particles below 600 microns diameter, while the powder metallurgy is more easily applied to particles in the region 500-2,000 microns diameter. Nukem is not in complete agreement with this statement.

The remark on particle size brings to the mind a general question :

is there an optimum size already fixed today

— on an economical point of view for fabrication,

— on the irradiation stability point of view?

And what would be the relative thickness of pyrolytic carbon to the kernel?

(*b*) Can the choice between oxide and carbide kernels affect the selection between dry or wet methods?

(*c*) Would not sol-gel be a requisite process if plutonium fuel had to be produced ?

As conclusion, it may be said, with General Atomic, that :

"There is at present no general recipe for preparing particles with a wide range of U/Th ratio. There is also no extensive production experience in the use of this process, which may mean that some of the disadvantages may not yet have been uncovered. More experience and development is necessary, but the "wet" processes seem to possess distinct advantages that will make them the preferred ones in the long run".

B. Coating

The coating applied on the fuel kernels must be of a low permeability to gaseous fission products and must also minimize the release of solid fission products. Pyrolytic carbon, which has a permeability coefficient (K) of about 10^{-12} cm^2/sec is an obvious choice as a diffusion barrier; however, it is relatively permeable to certain solid fission products such as caesium and strontium. One method to improve the overall retention is to place an interlayer of silicon carbide between layers of pyro-

lytic carbon. The silicon carbide provides a main diffusion barrier whilst the pyrolytic carbon is employed as a supplementary diffusion barrier and as a pressure vessel.

As indicated by Dragon, General Atomic and ORNL, the ability of the coatings developed to perform satisfactorily at high temperatures and for long burn-ups has been demonstrated in many experiments. Fractional Release Rate/Birth Rate of Krypton-88 during operation at 1,350-1,400° C can be as low as 10^{-5} to 10^{-6} and can be accounted for by the uranium contamination in the coatings as determined before irradiation by alpha-counting.

Pyrocarbon coatings are applied to the kernels by the pyrolysis of some hydrocarbon (mainly methane, due to its low cost and its purity), while the particles are suspended or agitated in some way. The dominant variables affecting coating structure are deposition temperature and the rate at which the coating is applied. Silicon carbide coatings are applied using methylchlorosilane.

The movement of particles is obtained :

(*a*) in a fluidized bed;
(*b*) in a rotating drum device;
(*c*) in a moving or vibrating bed.

(*a*) An extensive amount of work has been done on the properties and characteristics of the pyrocarbon deposited in a *fluidized bed.*

General Atomic says : "The process scales up nicely and except for criticality limitations, there is no reason why a very large coater could not be built to handle tens of kilograms at a time when thorium containing particles will be handled separately from the uranium containing particles".

ORNL presents some coating experiments on a fixed charge size and the same total gas-flow rate with variation of the temperature and the methane flow-rate. The results presented on Fig. 2 of the ORNL-report demonstrate that the properties of coatings deposited from methane can be controlled and that coatings having selected characteristics may be produced for optimization studies with respect to performance and the economics of the use of high coating deposition rates.

Porous inner layers can be deposited at very high rates from *acetylene* at relatively low temperatures (1,000-1,100° C).

One of the disadvantages of the fluidized bed is the normal use of large quantities of inert gas which has to be disposed of or regenerated. This might be avoided by the use of a technique recently tried by Nukem and which will be presented during the discussion.

(*b*) *Rotating drum coaters* :

General Atomic thinks that these coaters have the advantage of needing no addition of inert gas for levitation of the particles. They are, however, more complex mechanically and might present difficulties in remote operation.

Dragon feels :

— that the limitations are stemming directly from the need to keep soot formation to a minimum,

— that the deposition rate is relatively low,
— that the type of coating cannot be varied over wide limits.

Nukem will present during the discussion the result of their experience with this apparatus.

(*c*) *Moving or vibrating bed coaters.*

There is very little known at present on the operating characteristics of these coaters.

Discussion on coating.

1. — *Is silicon carbide coating necessary?*

The American experience tends to show that silicon carbide coating is not necessary if operation of the coated particles at a maximum temperature of 1,400° C is envisaged, if a R/B of 10^{-4} of the noble gases is considered acceptable for the primary circuit (10^{-5} to 10^{-6} is often obtained). It should be noted, however, that release on complete fuel elements might be different due to particle breakage during fabrication.

Limited data are available for the metallic fission products and long term irradiation results are necessary. The decision on the use of SiC should be based on

(i) the maximum temperature and time of fuel operation,
(ii) the economical ground (Dragon shows that the use of methylchlorosilane is an important factor in the price of coating),
(iii) the technique used for reprocessing : coated particles with silicon carbide must be broken before leaching and not simply oxidized (see Session II).

Dragon thinks that the problem of selecting SiC or not, cannot be resolved at the present time.

2. — *What type of coating must be applied?*

(*a*) Let us first remark that if extensive work has been done on coatings, no standard specification on coating structure, density and rate of deposition is available, except that a minimum average thickness of about 100 microns is necessary for a good irradiation behaviour. Does it mean that the specifications are proprietary information or that different procedures are available to produce "good" coatings? Long fast neutron irradiation doses might be an answer to this question and make the selection. Anyhow, duplex, triplex and multiplex layers have been tested with success up to now, but in thermal neutron fluxes.

(*b*) Should the first and/or second layer be used as plenum for fission gases? Depending on the answer, sintered or melted particles have to or can be used. If melted particles are chosen, on what model is the void volume calculated and what are the properties of the pyrolytic carbon used as reference?

The ORNL-3686 report : "A comparative evaluation of advanced converters", Jan. 65 page 47, describes a model :

"A simplified theoretical analysis has been used to predict dimensions of coated particles that may allow incorporation of the desired fuel loadings in the available fuel volume. This analysis is based on provision of a void volume as a porous inner layer (about 50 % dense) of the coating to accomodate swelling of the fuel core and to allow expansion of released fission gases so that internal pressures do not cause fracture of the outer impervious coatings".

It might be of interest during the discussion to precise the "optimistic" assumptions chosen for the study to enable to calculate the void volume depending on the burn-up to be reached.

3. — *Carbide versus oxides.*

Carbide particles have been successfully coated and irradiated. The precaution used in different manufacturing methods is to incorporate an excess carbon (10-20 %) in the kernels to avoid any coating dissolution at the operating temperature.

Retention of the structural integrity of those particles under severe irradiation conditions led to serious consideration of utilizing carbon coated oxide fuel particles for HTGR applications.

ORNL states : "A matter of possible immediate concern is the basic thermodynamic instability of oxide fuel compounds and carbon under both fabrication and operational conditions; however, this concern may not have basis in practice if it can be demonstrated that the low-permeability coatings can be applied satisfactorily and that they will retain their integrity so as to withstand the equilibrium partial pressures of the reaction product gases".

The main technical questions with such particles are :

(*a*) the effects subsequent to particle breakage : the reduction of oxides to carbides with evolution of CO, which might not lead to the important corrosion problem mentioned by Dragon;

(*b*) the effects of long term fast neutron irradiation which are anyhow of importance for all coatings, independent of the kernel nature;

(*c*) whether oxide fuel kernels have significantly different retention properties compared with carbide kernels.

The use of oxides might be of particular importance when using plutonium as the carbide use introduces technological difficulties due to its vaporisation characteristics.

The influence on cost will be discussed later.

Oak Ridge presents some results obtained on sol-gel, theoretically dense, spherical $(Th,U)O_2$ particles. Temperatures of only about 1,200° C are required for final densification. The pyrolytic carbon coatings were applied without difficulty.

Thermal stability tests indicate that adequately thick coatings (greater than about 60 μ for the extreme temperatures) are apparently required to withstand the equilibrium CO pressures developed during elevated temperature exposures in vacuum

(about 1,800° C for 4 hrs). It was noted, however, that two batches of particles deposited from methane to average thicknesses of 43 to 50 microns survived treatments at 2,000° C for 50 hrs and at 2,200° C for 2 hrs with no detectable damage. The role of coating structure is being investigated.

The irradiation test results : ORNL report, page 16 : "The highest burn-up demonstrated for base coated uranium oxide particles is 8 at % heavy metal at 1,400° C in a sweep capsule... By comparison with results from other tests in the same facility, the fission gas release has been constant and very low (2×10^{-7} R/B for Kr^{88})".

Further experiments are necessary under both thermal and fast neutron fluxes

III. — Fuel element concepts

As Dragon points out, there are today divergences in the design of HTGR fuel elements which are likely to have a significant effect on cost. While General Atomic describes rapidly its Peach Bottom fuel element, three reports present results on 6-cm fuel spheres development, either for the AVR loading or for the THTR association programme within the Euratom countries (THTR = Thorium Hochtemperaturreaktor).

The fuel spheres in a pebble bed reactor are subjected to impact loading, steam oxidation and abrasive action during reactor operation and therefore it is mandatory that the coated fuel particles are protected by an unfueled graphite shell. This shell might be either molded or machined from fully graphitized material.

Limited ORNL data indicate that all-molded elements are superior to machined shell elements, although they lose some impact resistance upon thermal cycling in vacuum. ORNL presents some *irradiation test performance* results of different spheres irradiated in the ORR poolside facility. Relative fractional release of noble gases shows little difference between the molded and machined shells. A somewhat wider variation is apparent for different molded and machined shells. This probably indicates variation in the effectiveness of bonding between the matrix and the shell, bonding which represents an important problem area.

This is further shown by *crushing strength measurements* : the strength of the fueled spheres is quite dependent upon the mechanical support provided by the matrix. Comparisons of irradiated and unirradiated spheres show little difference for the longer exposures obtained, up to 6×10^{20} neutrons/cm^2 (E greater than 0,18 MeV). Concern still exists that increased shrinkage may be experienced for longer exposures or a harder neutron spectrum.

The release of solid fission products has been studied, but no relationship has been established to correlate these observations with the noble gas release or the fuel element design. It is apparent that this important property of the fuel requires additional investigation.

The thermal conductivity of fueled graphite spheres as affected by irradiation varies significantly with temperature, as calculated from irradiation results (see

Fig. 8 - ORNL report). Little variation with irradiation dose was noted up to 10^{20} neutrons/cm^2 (E greater than 0,18 MeV).

Let us review now different fuel element configurations :

(*a*) *the AVR first core loading.*

This fuel element is manufactured by Carbon Products Division of UCC in Lawrenceburg (Tenn-USA). The coated particles are uniformly dispersed in a graphite flour matrix, filling the whole sphere interior by a mold injection and baking technique. For high fuel loadings, considerable care must be exercised during pressing to prevent damage to the coatings. Further details will be given by Dr. Eatherly during the discussion.

(*b*) *Nukem developed fuel elements* (see Fig. 1 of the Nukem report).

b-1

Fuel elements with loose coated particles.

Two types of fuel elements with loose coated particles have been considered. The use of loose particles should be in principle one of the cheapest methods of consolidation.

1. *The annular gap element* (Fig. 1*b*) (25 mm ⌀, 25 mm height, 2 mm thick), relatively easy to produce, but is not suitable for normal AVR or THTR elements because the volume of the gap is too small to accomodate coated particles with a sufficient heavy metal content. If the gap width is increased, the central temperature becomes too high. The thermal conductivity of a loose particle layer in reactor conditions has been extrapolated from measurements in helium up to 700° C, the value is 0.005-0.01 cal/cm. sec. °C.

Several of these elements have been successfully tested in Risø (Denmark) using Dragon coated particles with burn-ups up to 21 % of the U^{235}.

2. A new type with a spherical gap, proposed and examined by the Ringsdorff werke, Bad Godesberg, is shown in Fig. 1*c*. The gap is produced by incorporating pieces of volatilisable material during the moulding of the spheres; then, the plastic material is volatilised and the production process finished by carbonising and graphitising the moulded spheres.

b-2

Hollow sphere fuel elements (Fig. 1*a*).

These elements have a thin shell of about 10 mm thickness. Systematic comparison of the physical and chemical properties of different graphites is underway in the THTR program, moulded graphites being preferred to extruded ones. Dropping strength of full and hollow spheres is particularly studied, though this property was until recently unknown to graphite producers and users.

The coated particles are arranged in a thin layer of about 1-2 mm adjacent to the internal wall of the hollow spheres; a lower in-pile temperature of the hottest fuel particles can be expected.

The wallpaper type fuel elements are produced by filling a slurry of the coated particles with graphite powder, binder and solvent into the hollow spheres. The

spheres are tumbled in order to produce an even layer of particles while air is blown inside the spheres to dry the slurry. Particles containing up to 11.5 g of heavy metal can be inserted in one tumbling step. The inside of the sphere is filled with natural graphite powder without any binder. A plug is screwed in with a binder. Cracking is made at 1,000° C in argon and at 1,500° C in vacuum. No particles are damaged during fabrication. The first irradiation results are favorable.

b-3

Synthetic fuel element (Figures 1*d* et 1*e*).

The graphite matrix of the synthetic fuel element differs from electro-graphite mainly because the final heat treatment is maximum at 1,700° C to avoid metallic diffusion in the coatings.

The graphite body is formed from a mixture of graphite powder and binder; as this binder is only carbonized and not graphitized, it is desirable to use as little binder as possible. Natural graphite powder is used as grist together with some artificial graphite powder and sometimes a small percentage of carbon black.

A semi-hydrostatic forming process with a rubber insert die is used to limit particle breakage.

The development is not yet completed : systematic irradiation and corrosion tests must be performed. But several types of balls produced from different mixtures show very promising properties. Nukem states that "this fuel element might be expected to be cheaper than the types with machined electrographite shell in the primary fabrication; it is difficult to predict whether this will still be true in the whole fuel cycle, including reprocessing and refabrication with active fuel".

IV. — Fabrication cost

Let us first regret that very limited information has been given on coated particles or fuel element fabrication costs in the papers assigned to Session I.

— GA feels that fuel for a 1,000 MWe capacity High Temperature Gas-Cooled Reactor can be fabricated in the next few years for about 200 dollars/kg (of heavy metal) for the particles, plus about 500 dollars per element for graphite and assembly.
— ORNL refers to the Lotts and al paper, assigned to Session II.
— Nukem does not present any financial estimation.
— The only detailed financial evaluation has been made by Dragon in an extrapolation of its experience gained during the manufacture of fuel for the first charge of the Dragon Experiment. It is impossible in a short presentation to present all the data included in the paper; indication of the main hypothesis and of the main questions raised might lead to an interesting discussion.

Dragon assumes that for the first generation of HTGR *a once through* fuel cycle will be used. The upper limit for an initial nuclear power programme can be considered to be set by the level at which reprocessing fo fuel would become economic and this is deduced to be in excess of 2,000 MW(e) installed capacity.

A fuel fabrication capacity has been chosen with a design output of 1,135 kg U^{235}/year (about 40 kg heavy metal per day with Th/U = 10), capable of fuelling a 2,000 MW(e) [5,000 MW(th)] programme consisting of four reactors, one station being constructed each year.

For the case which has been chosen, with a Th : U^{235} atomic ratio of 10 : 1 (N = 10), the initial feed for each station is assumed to be 730 kg U^{235}, the annual make-up thereafter being 270 kg/station.

Main technical assumptions :

— 5 % recoverable rejects, 1 % irrecoverable loss;
— particle preparation can be automated;
— output of sintered kernels in a single batch continuous furnace ;
— output of coated particles with two fluidized bed furnaces with 6 inch diameter reactors (plus one spare). (GA uses 5 inches furnaces, 7 kg particle capacity).
— four process cells in the active area.

Main economic assumptions :

— The fuel production unit would be part of a larger, but not necessarily nuclear complex (which is a more logical assumption than the ORNL one of a complete independent facility);
— the production will be carried out for 300 days per annum on a 24 hours basis, using 4 shifts to run a 3 shift system (ORNL 260 days per year);
— a preproduction run will have costs equivalent to 6 months running cost without charge for the U^{235};
— no allowance for profit has been made (factory cost versus selling price).

Results :

The estimated fabrication cost for coated particles is (excluding enriched uranium) :

\$1,300/kg-U^{235} or \$120/kg-heavy metal or \$59/kg-coated particles.

Discussion : (refer to Table 5 of the Dragon report).

(*a*) Less than 50 % of the cost depends on the chosen fabrication route.
(*b*) The most expensive part of the process is the *coating* and the fruitful area for economy is in the raw materials, almost all the cost being incurred in the supply of gas and silane to the fluidized bed furnaces.

The argon is used on a once-through basis and
— methods of recirculation are worthy of investigation,
— methods of eliminating completely the argon :
  — by a replacement by hydrogen, nitrogen or any cheaper gas mixture,
  — by using a rotating furnace which suppresses all fluidizing gas (gain 20 %).

Dragon concludes, however, that the tumbing bed method is likely to be a slower process with less output per unit volume of furnace hot zone. Suppression of methylchlorosilane, which is about ten times more expensive than methane per unit volume of coating, would be another economy.

(*c*) *Conversion from uranium hexafluoride to* UO_2 is the next most expensive system. However, the figure quoted is a buying price and a factory making its own conversion might economize on the operation.

Further studies on this conversion or methods of proceeding directly from UF4 to carbide might be of interest.

(*d*) *Thoria and carbon* : a small economy can be obtained on the thorium price if the figure given in the ORNL report is adopted.

(*e*) *Irrecoverable losses* : on its previous experience, Dragon feels that 1 % loss is a more realistic figure than 0.2 % adopted in the ORNL-3686 study.

Integrating all possible economies, the price of coated particles goes down to (see table 6 of the Dragen report).

$$\$1{,}160/\text{kg } U^{235}, \text{ saving } 10.5\ \%$$

Two further remarks are necessary :

(*a*) Dragon is not able to compare the kernel preparation and sintering costs to other possible variations in the methods; those costs represent :

$$\frac{48.1 + 32.4}{417.4} = \frac{80.5}{417.4} = 20\ \% \text{ of the total.}$$

Might it be possible to introduce some savings in this field (Nukem has tested other methods) ?

(*b*) Dragon has considered carbides only; Lotts in his paper discussed in Session II, estimates that reduction in fuel fabrication costs of greater than 15 % can be achieved if oxide rather than carbide particles are used, the economy varying with the production size.

Effect of variation of Th/U^{235} *ratio on cost* : it shows a linear dependance with N value up to $N = 10$.

The cost of consolidating coated particles into fuel elements is studied with the following hypothesis :

— the consolidation is assumed to be carried out in an extension of the coated particle production building;

— a prismatic form of core arrangement is envisaged.

The *total fabrication* cost is (excluding enriched uranium) \$235/kg heavy metal or \$2,600/kg U^{235}. The use of free coated particles reduces the cost to \$2,100/kg U^{235} or \$195/kg heavy metal.

Strong points in the use of free coated particles are minimization of mechanical damage to the coating, a high fuel density achieved as well as reduction in the reprocessing cost. In the balance must be placed the lower heat transfer characteristics and the risk of coated particles release in the primary circuit.

In the fuel element fabrication, the graphite blocks for the prismatic core arrangement dominate the cost of consolidation. Efficient use of the graphite should be one of the dominant design features.

U^{235} inventory : about half of the financial charges derive from the U^{235} inventory in the fabrication plant :

In conclusion : different improvements in the coatings and fuel element fabrication might lead to decreases in the actual fabrication costs.

SESSION I : DISCUSSION

Chairman : P. CAPRIOGLIO (Euratom)

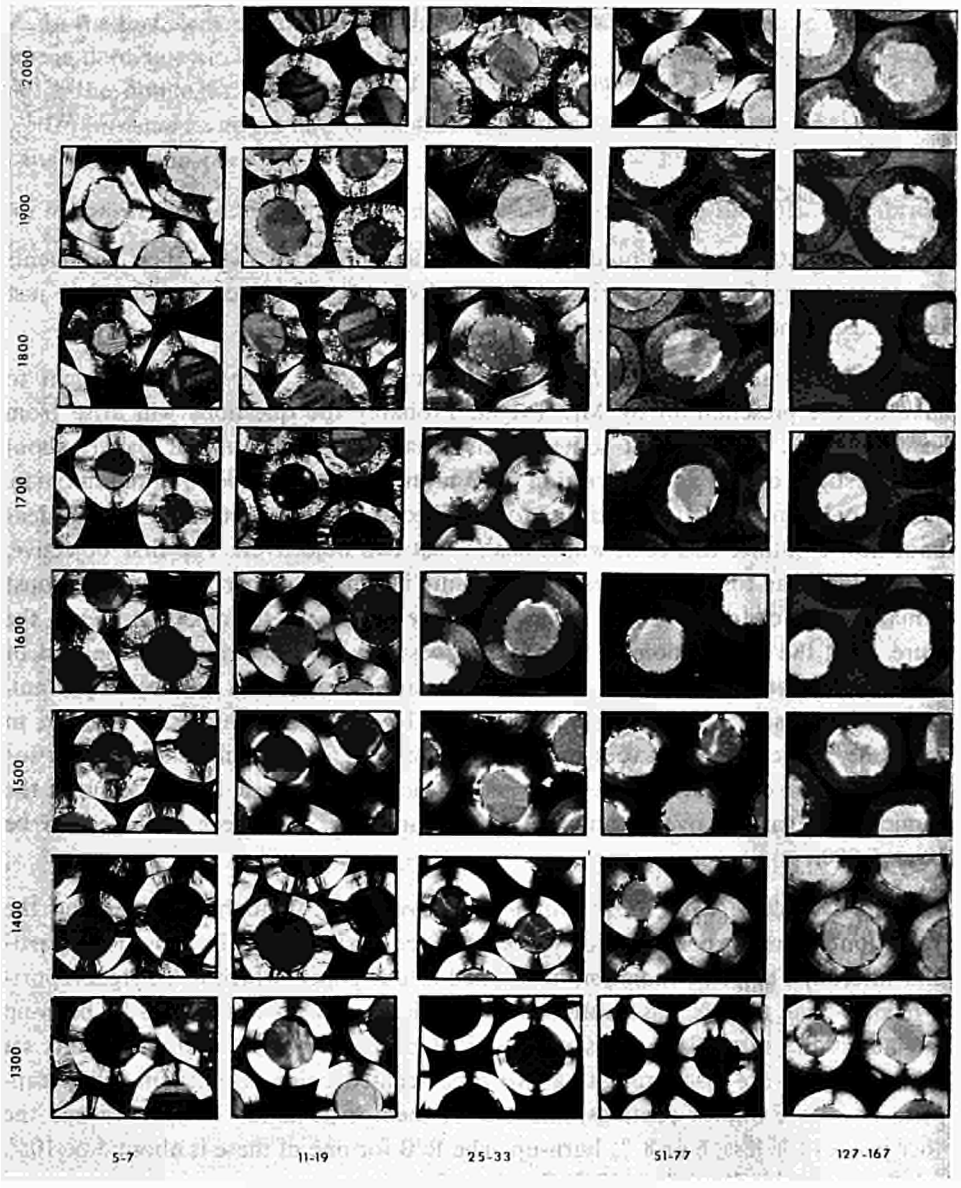
1. — AUTHORS' COMMENTS.

Dr. CAPRIOGLIO : I should like to ask the authors to make their comments and present any further remark or serious divergency they see in what has just been said by the rapporteur.

Mr. HOUDAILLE *(Dragon Project)* : I don't think I have much comment to make on the presentation by Mr. Tytgat. Probably the questions will arise from the discussions. I know that some criticism has been made of our paper, about these questions of loss and conversion from uranium hexafluoride to uranium oxide. There is only one thing I should like to add, because it may give some wrong ideas about how to utilize this cost, which has in fact two objectives. The first objective, in my mind, was to give an idea of the points in which further development must be made in order to save some money when we shall have to make the fuel in the future. And the second point is that in this cost we have incorporated the cost of raw material for graphite and for thorium. And I think, this can be important, because I saw some studies of fuel cycle cost based on the physics of reactor, in which the fabrication cost does not include the cost for thorium and the cost for graphite. To give an idea of the influence of the graphite cost, I must say that the atomic ratio, carbon over uranium-235, in our paper has been assumed to be about 5,000.

Mr. TRAUGER *(Oak Ridge National Laboratory)* : I might comment on the irradiation testing of coated oxide particles, since this seems to be a point of particular interest. The one irradiation mentioned in our paper, which Mr. Tytgat reported having gone to 8 % has continued in irradiation and is now at 18 % burn-up with about the same fission gas release, R/B of 2×10^{-7}. In addition to this set of particles, we are also irradiating two other coated oxide particles in sweep capsules. These are duplex coatings, and one of them is at about 10 % burn-up, the other just a little less, 6 or 8 % burn-up. The R/B for one of these is about 5×10^{-7}, and the other is about 1×10^{-7}. So the release rate experience for krypton and xenon is very favourable thus far to these burn-ups. It also is interesting to note that there is at least some correlation between the alpha activity observed on the surface of the particles and the R/B for these particles.

In our paper, we have shown the characterization of coated particles in a series of particles which were prepared for this purpose, in which the density, the crystallite size and the anisotropy were measured, and I thought it would be of interest to show two slides. On the first one (figure 2 of the paper), see p. 31, we have plotted the deposition temperature versus the methane flow rate, showing the various



Methane flow rate, $Cm^3/Min\text{-}Cm^2 \times 10^{-2}$

Effect of deposition conditions on microstructure of pyrolytic-carbon coated UC_2 as polished, polarized light.

properties of the coatings as determined from measurements on small discs that were coated with the particles. And you will note that the density varies as shown here, one can obtain high densities either at high or low coating rates, and high or low temperatures, and the crystallite size varies quite uniformly. If you will retain

the image of this slide showing the anisotropy factors and the high anisotropy in this region, and the general shape of these contours, the next slide (1) "Effect of deposition conditions on microstructure of pyrolytic-carbon coated UC_2" will show a composite photomicrograph of particles of single layers under polarized light and you will notice the Maltese cross on the particles having higher anisotropy. The Maltese cross under polarized light is characteristic of particles having higher anisotropy although its absence does not necessarily indicate an isotropic particle. And if you look carefully at the slide, you will see that the regions of high anisotropy shown on the previous slide are evident. You can pick out the contour lines of the previous slide, showing that at least there is some correlation between the properties measured on the little discs and the particles themselves.

We are continuing these studies, making duplex particles for irradiation testing. These particles will be irradiated in large capsules containing several groups of particles irradiated to identical conditions for comparison of the irradiation properties by post-irradiation examination. These are screening tests and those which look good in post-irradiation examination will be tested in sweep capsules, to high burn-ups following our normal testing procedure.

Dr. CAPRIOGLIO : Mr. Trauger has given us very interesting information in particular on the behaviour of these oxide fuels, and I think we have to come back to this in the discussion.

Dr. RAGONE *(General Atomic)* : I would like to second Mr. Trauger's emphasis on the properties of the coatings and the measurement of anisotropy factor and density as the variables that are important in the performance of these coated particles in the reactor. There is some data on these factors in our paper as well. — It is our feeling that through a consideration of these properties, we will be able to optimize the coating thicknesses and processing for production of retentive coated particles.

There is one comment I have on the silicon carbide. We don't, at least in our experience, consider that the silicon carbide is the layer that retains the gaseous fission products. We think of the silicon carbide as being put there mainly to retain the metallic fission products. We would like to offer the idea that, even without the silicon carbide layer, the metallic fission products will be held up — kept out of the primary coolant — by the graphite in the fuel element itself. That is the metallic fission products will be adsorbed on the graphite of the fuel element.

I have one more short addition. In my paper I did quote something about the GAIL-loop, the full-diameter section of a fuel element that we are irradiating in the GETR. It contains loose coated particles in a telephone dial arrangement that is shown in the last figure of my paper. This fuel element was put into the loop

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(1) This slide will be included in the paper by R. L. Beatty *et al.* "Pyrolytic carbon coatings on ceramic fuel particles" to be published in *Nuclear Applications.*

last summer and is now at 30,000 MW/days per ton peak burn-up, and is performing very well, i.e. the R/B ratio for gaseous fission products is between 10^{-6} and 10^{-5} at temperatures up to 1,500° C.

Dr. CAPRIOGLIO : I think it is now the moment to ask Dr. Eatherly to make his contribution, that he has promised us on the fabrication of the AVR fuel at Union Carbide.

Dr. EATHERLY *(Union Carbide)* : I sincerely apologize that our paper is not in your hands; we used our best efforts to get it here, but we figured without the efficiency of the Belgian customs agents.

I am only going to review the central part of our paper, starting page 5, paragraph III. (1).

Graphite compounds. For the mixtures we are using in the AVR balls, if left unconstrained, we are using expansions of the order of 40 to 50 %. Of course, in the graphite shell, this remains constrained by the shell, and after the temperature treatment is complete, the residual strains are removed. It is quite easy, incidentally, by adding a little too much bisulphate unto the matrix, to blow rupture the ATJ balls without any difficulty.

Irradiation behaviour. To date, the irradiation behaviour of these AVR balls has been determined entirely by Oak Ridge National Laboratory. Balls have been irradiated at temperatures approaching that of the AVR reactor, at burn-ups of the order of 5 to 6 %. The temperature gradients have been measured in this type of ball. The thermal conductivity seems to be excellent. Because the temperature gradients do not change during irradiation, there is apparently no indication that the matrix shrinks away from the shell as a result of irradiation damage. We did not anticipate that this would happen, since the natural graphite that we use as a basis for the residue compounds should expand during irradiation and thus further stabilize the matrix against any contraction during damage.

Economics. There is no question in our minds that the selling price of 23 dollars per ball is excessive. On this order, and on other orders, that we have processed to date, we produce about the same weight of paper as we produce of uranium. Certainly, in the early stages of the fuel element work, overcaution is necessary, but there is no doubt in our own minds that the price of this type of element or any other high-temperature gas-filled reactor element can be brought down considerably. In the United States, we have seen this happen in the last few years with coated particles where the price has dropped by a factor of 2 or 3; it is still dropping, so that we have every confidence in the world that the price of the fuel element fabrication for this type of reactor will come down considerably, and the future of gas-cooled reactors is very bright.

I have a few slides of our production facilities. Since we are not part of the Euratom group, I thought you might be interested. This is a photograph of our

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(1) Only the comments not included in Dr. Eatherly's paper have been reported.

facility in Lawrenceburg, Tennessee; it has a capacity of about 200 kg of uranium per month, operating as a fabrication plant for the AVR fuel element. We spheroidize our particles in a plasma torch at a temperature of about 15,000° C. The particles are dropped in at the top, are collected at the bottom and removed within a glove box to protect the thorium against any possible oxidation.

The coating furnaces are a five-inch diameter graphite induction heated vessel. The shell that surrounds them is simply a protective device in case we have a water leak from the induction coils, to prevent any injury to the operators.

Dr. CAPRIOGLIO : I think that our main problem now is to give some order to the discussion and take the items one by one in order to avoid dispersion. So I would suggest that we take first of all the questions that have been raised by Mr. Tytgat in his presentation, going from the inside of the coated particle towards the complete fuel element. We could talk first of all of the technical aspects and then end up with the final juice which is the cost of all this.

2. — USE OF OXIDE PARTICLES.

I think, one important question is, for all of us, the question of using *oxide particles*. I have myself for a long time been convinced, perhaps on purely irrational

grounds, that this was a very good idea, and I am very pleased to see that the first irradiation experiments that are being carried out at Oak Ridge seem to be very promising, but I should very much like to see how various people working in this field react, and finally what the possible technical and economic advantages can be. Between others, there is one thing that I should like to see discussed : What is the high-temperature behaviour of these particles? This is because I got the impression that, almost paradoxically, pyrolytic carbon coated oxide particles could stand higher temperatures than carbide particles, in spite of the compatibility problem of the oxide with the pyrolytic carbon. I should very much like to see if anybody has done any work on very high temperatures on oxide particles.

Mr. HUDDLE *(Dragon Project)* : I think the oxide/carbide controversy is one of the major problems we must sort out in the initial stages of coated particle fuel development. An important aspect of high-temperature reactors is that of corrosion and mass transfer of the core, because although we look on designs where graphite is replaceable, the cost of replacing that graphite is not insignificant. Now, we know that the capital cost of a large purification plant is high, and therefore it seems to me a fundamentally bad philosophy, (unless there are other inherent advantages of the oxide route) to put in something that on long-term running such as 5-10 years, — the sort of times we are thinking about — could add to the hazards of coolant impurity i.e. oxygen in the reactor system. As an ex-corrosion man, it seems to me that to operate with a system that is thermodynamically unstable — particularly with a broken particle which reacts very quickly — is fundamentally bad philosophy. Regarding the temperature, we have had carbide fuel operating in the Studsvik irradiation facility up to 1,900° C and it appeared to behave very satisfactorily.

The other point, regarding costs : there is in one of the papers a statement indicating that oxide fuel is very, very much cheaper than carbide fuel. Well, our experience does not agree with this because the only difference in a production unit is the difference of cost of heating a green particle to transform it to carbide, as opposed to heating a green particle — the oxide — to transform it to a stable oxide. I would think that the additional cost is insignificant, at a guess perhaps 0.1 %; I honestly do not think there is any more in it. I feel therefore that, in the oxide field, we are in the very early stages of development, and to try to give an unbiased and worthwhile opinion at this stage is, I think, looking into a crystal ball.

Dr. RAGONE *(General Atomic)* : As far as cost of production is concerned, I think I would take issue with your estimate of less than 1 % difference in cost between oxide and carbide. There are many advantages of dealing with an oxide, not the least of which is the avoidance of the hydrolysis problem for the thorium-dicarbide.

Oxides can be handled easily in an open room, and can be stored easily, giving a greater flexibility of production scheduling. These advantages, perhaps each one of them small, add up to what I think will be a considerable saving in production.

So I think, your estimate of less than 1 % saving by going the oxide route is too low.

Second, on the release of oxygen from fractured particles, oxide particles can be heated to quite high temperatures without fracturing. You seem to imply that the fracturing of oxide particles will be a routine thing in the reactor, and of course we do not expect that. That is, the release of oxygen from these particles will be under unusual conditions if it does occur, and at that point the plant would be shut down and the normal clean-up equipment would be used to remove any oxygen from the circuit. I do feel that you will get a release of oxygen from these particles through the pyrolytic carbon and have to increase the size of your oxygen clean-up system because of the presence of oxides in the reactor.

Dr. SHEPHERD *(Dragon Project)* : I do not think one should be too preoccupied with the cost of making these particles. If you accept the figures in our Dragon paper, then it is apparent that the cost of fabrication is already very low compared with the cost of uranium-235. And that being the case, I think one has to be concerned with producing the best product; because it only requires a small margin of difference in the performance of the reactor to offset any differences of cost that you might get in trying to make the product a little cheaper. For example, if we halved the coating cost given in our paper at the expense of losing a half a percent on the conversion efficiency of the system, we would have offset the lower production cost. So one has to be very careful not to try and go too far in reducing costs at the expense of obtaining a poorer product.

I raised the point particularly to answer the question of whether silicon carbide is better than pure pyrocarbon. There seems to be no doubt at the moment that silicon carbide does stop the diffusion of metal fission products. and I think this is a sufficient reason for having it, even though it may be marginally more expensive to put on as coating. Some of these metal fission products can catalyse corrosion, and this is a very important point which cannot be ignored. It is true that the graphite will, to a large extent, hold up these substances, but it holds them up in a place where they can increase corrosion rates, and this could be serious. Unless we can prove that pyrocarbon can be developed to the stage where it will be as good as the composite coatings, we are not inclined to reject it, because we think we are not losing anything in the slight increase in production cost.

Dr. CAPRIOGLIO : Many thanks, Dr. Shepherd. I think the silicon carbide question is another important one that we should discuss further on. Before going into this subject, however, I think we should ask what the feeling of the Oak Ridge and Union Carbide and Nukem people on this oxide problem is. I think, it is worth insisting on it because, apart from the question of fabrication cost, one has to think also of the problem of refabrication after reprocessing, and it could very well be that what seems to be a marginal advantage for just fabricating these fuel elements for the first time, can become a serious one, perhaps, in refabrication out of contact, when reprocessing would be a more economic proposal than a once-through cycle. Before asking whether Dragon has any comment on this, I would like to see what

the feeling is on the Oak Ridge, Union Carbide and Nukem side on the oxide route.

Dr. EATHERLY *(Union Carbide)* : I can only emphasize, what Dr. Ragone said already. In the last six weeks or so, we have swung our plant over from pure uranium to uranium-thorium system. The problems that we have run into are more than we anticipated. The principal problem has been the hygroscopic reaction of thorium carbide with water vapour or oxygen; not only has this meant a considerable tightening up of every piece of equipment we have, but it has pretty well destroyed a lot of the scheduling — ease of scheduling — that we had on pure uranium. Having seen what this has done to our costs, we can see, at least in our process, a tremendous advantage to the oxide fuel. We also keep in the back of our minds the question that one of these days we will have to face up too, and that is re-processing. And seeing these difficulties in our original fabrication, we shudder when we think of doing it as a remote step. While I can only express our point of view, we feel that the oxide represents a considerably easier material to fabricate and therefore will be much cheaper.

Mr. LOTTS *(Oak Ridge National Laboratory)* : I would like to point out that not only does one have the cost of converting the oxide to carbide, but also the problem of maintaining an inert atmosphere during a substantial number of operations, if one is handling carbide. Therefore, that is a substantial cost contributor. We have estimated in a paper that is in the second session today that the cost of conversion would range from approximately 15 % to 5 % if one is fabricating in an extremely large fabrication plant.

Mr. DOUGLAS *(Oak Ridge National Laboratory)* : I would like to add to the comments that Mr. Lotts has made. If one considers the technical performance of an oxide particle in a reactor, I think that certainly in our Laboratory we still have an open mind as to whether or not such a concept is an acceptable fuel system. We also appreciate the fact that the Dragon people had to make decisions many years ago as to what concept to use. In subsequent years, new information has developed and, thus, I think that our work on the oxide is perhaps pointed towards an advanced system. With the limited available data on irradiation, it is certainly too early to make any concrete decisions at this time.

In my opinion Mr. Huddle is unduly alarmed about the question of the thermodynamic instability of a carbon coating on an oxide particle. At the proposed operating temperatures in Dragon for example the kinetics of the system would not permit the conversion of an oxide to a carbide as all of us who have attempted to make uranium dicarbide in a carbon bed can attest. At temperatures above 1,700° C one does, in fact, convert the oxide to a carbide if a defect in the coating occurs. One experiment indicates that no unexpected amount of fission products or fission gases are released as a result of this conversion.

Dr. WIRTHS *(Nukem)* : As a production man I should like to say that all that Dr. Ragone and Dr. Eatherly have said also holds for us. Economics can be considerably improved by mixing of oxide particles: but one should not forget that the cost of producing the particles depends far more on the quantity. The pity of it is that today the quantities to be produced are negligible, and so it is by far too expensive.

Another point which has not been mentioned today is the diameter of the particles. I say that the diameter of the particles is increasing and this clearly improves the economics if we can make the particles bigger than today.

Thinking of re-fabrication, especially remote handling, I think that the advantage of the oxide particle is bigger, still bigger than in the first step where you can produce in simple glove boxes.

Dr. CAPRIOGLIO : I think, Roy, that this is a sort of lonely fight, so I suppose that I should again give the microphone to you.

Mr. HUDDLE *(Dragon Project)* : This is the sort of contest in which I am very, very happy to participate. The only thing that causes me any concern of losing is, if some so far unknown subtle effect of oxide gives rise to a superior product. However, we are trying oxide fuel in the Reactor Experiment : in fact, in our initial proposals for the second charge, oxides had a major share of the programme.

Now, there are two things I would like to mention regarding the thorium problem. In the very early days of coated particle fuels, as some of my staff remember well, I refused to allow them to work on plain uranium carbide. We went straight to thorium and — as a result — solved the problems at the outset without great difficulty. In the production of the first charge, the development people had adequate experience and we had no trouble whatsoever regarding the handling of thorium dicarbide. However, I can well understand Dr. Eatherly's problems in transferring from a system that is perfectly satisfactory for uranium dicarbide to one for thorium. For production however, I feel, the whole problem can be eliminated by a very simple double-furnace unit where you put in your green particles in the top furnace, heat treat them to carbide, transfer them into the fluidizing furnace, coat them and take them out into air. There are no special inert atmospheres, except those that are involved in the fluidizing. You heat treat in vacuum and fluidize in the normal way. This is the system we have in mind.

Another point that has been brought up is regarding reprocessing. Now, two years ago, I honestly felt that to deal with a powder metallurgy route involving carbides in a concrete hot cell was almost impossible. Now we feel, it is a relatively simple matter and would have no fears in having to build such a unit. We already have considerable experience from producing the first charge of the reactor experiment, and now, in our work on plutonium, we are making what I consider the first step towards remote handling : that is, putting our own methods, the powder me-

tallurgy route (and the sol-gel route) in a plutonium glove box line. From what I can see, we are not running into any serious difficulties in this respect.

Regarding Dr. Douglas' comment on the release from oxides after the coating has fractured, the fact that they are no worse than carbide is, I think, irrelevant, since the release from a broken particle is so great in comparison to an unbroken one. What worries me more is the fact that you have released oxygen into the system, carrying with it what I consider rather nasty problems (mainly because their extent is not known) of corrosion and mass transfer.

Dr. CAPRIOGLIO : I should like to ask at this point Mr. Valette to tell me what his feelings are concerning this problem of oxygen in the primary circuit coming from oxide particles with broken coatings.

Mr. VALETTE *(Euratom/THTR)* : I was ready to back Roy Huddle on this point of reactivity of the kernel and the coated particle, mainly based on information which was obtained from Dragon staff. I was told that some tests were performed on coated particles in the presence of water vapour. After the equivalent of 40,000 hour/micro-atmosphere (which is the equivalent of 1,000 hours in 1 vpm, assuming a 40 atmosphere pressure in the helium) in most of the coated particles using only pyro-carbon, 1 % of the coatings failed in the best of the coated particles, and in the worst coated particles, 20 % of the coatings failed. So this is the result to be expected if coated particles are at 1,000° C in presence of 1 vpm after 1,000 hours. But no failure has been observed with silicon carbide coated particles. So this is the first point.

The second point of oxygen released from the kernel itself — I have not made any exact calculations, but I do not feel that this would be a real problem, because the rate at which this oxygen would be released, would be progressive — I do not expect that all the kernels will break suddenly. It will not be comparable with the acceptable water in-leakage in the core.

Dr. CAPRIOGLIO : If I understand you properly, you agree with Roy Huddle that silicon carbide is a good thing, but you do not agree that the oxide particles are a problem?

Mr. VALETTE *(Euratom/THTR)* : I would say so, when I assume that this destruction of coated particles will only come progressively. If, of course, all the coated particles break at the same time, we would be in trouble; but if you have progressive breakage, it will not be worse than carbides.

Prof. SCHULTEN *(KFA/THTR)* : I think it is very easy to make an estimation about an oxygen content in the reactor, which is coming from the uranium or thorium oxide, because very big systems will have about 10 tons of heavy material, something like this; from this there will be, let's say, 10 % of oxygen, that means, there will be 1 ton of oxygen in this system. And let's take the case that 10 % of coated particles are broken, then we have about 100 kg of oxygen in the reactor,

and that means that is in the same order as there is oxygen in the form of water in graphite and as impurity in the reactor. And these 100 kg of oxygen are coming in, let's say, one or two years, step by step, so that the contribution of this oxygen is rather small.

Dr. RAGONE *(General Atomic)* : This question of oxygen coming out of particles — let's argue in the following way : that the oxygen will not come out of a particle unless it breaks. I think that Roy Huddle and I both accept that. But if the particles do break, then you have two kinds of problem : one is the oxygen release and the second is the fission product release. I think that the fission product release is so much more serious that the reactor would be shut down on that basis far sooner than it would on an oxygen release basis. I have just made a small calculation for a reactor of our kind, the HTGR. If we broke a million coated particles, we would release about 40 standard litres of oxygen. This would amount to something like 4 parts per million of oxygen in the system, that is if it is distributed uniformly throughout all of the gas. But the release of fission products would I think be far more serious and we would tend to shut down the reactor far sooner on that basis than this increase of 4 parts per million in oxygen.

Dr. CAPRIOGLIO : Well, I do not think that it is likely that we can go much further, and I am not hoping to convince anybody today, so I think we can leave this. I suppose that everything that had to be said, has been said.

3. — SOL-GEL METHODS.

I should like to take the next point which seems to be a sourse of controversy and very different opinions. It is the use of the *sol-gel method* of making the particles, as compared with the powder metallurgical methods. And again I think it is good to start with Dragon, and I am wondering if Roy Huddle or anybody from Dragon would like to comment. I would suggest that only the principles of making use of sol-gel methods versus powder metallurgical method should be discussed, rather than details of the methods that are being used.

Mr. HUDDLE *(Dragon Project)* : When I first heard of the sol-gel process from the late Dr. Dean of Oak Ridge it seemed that here was an ideal process, and we were very keen on it, since there is no doubt that it has many advantages. However, as far as I can see, no one has any significant experience of enriched sol-gel fuel production where irrecoverable losses can be checked. I think we have not yet reached the stage where a true comparison can be made, because in our experience, the powder metallurgy route is equally suitable to the sol-gel route for remote fabrication. Once you are experienced, such as with a violin, you make good music : the same applies to the powder route and its product. I think therefore that we cannot yet make judgement. The primary factor surely, as Dr. Shepherd mentioned, is the perfection of the particle, and in this lies the great advantage of the sol-gel route in producing a particle of excellent shape. Apart from technical perfection,

I think a most important factor in fabrication is the effect of losses. Furthermore, we feel it is easier to maintain a remote fabrication plant, employing the powder metallurgy route as we envisage it now, than to maintain a sol-gel route. There are many problems in "sol-gel" that have yet to be solved; undoubtedly they will be solved, but again I think at this stage we must await experiments which enable us to compare the two side by side. We have at Winfrith a notice in the Dragon Fuel Element Development Laboratory which reads "One experimental result is worth a thousand expert opinions". I think it is relevant.

Dr. RAGONE *(General Atomic)* : Concerning the business of remote operation, I cannot help but ask Mr. Huddle if he has ever tried to finger a violin in a hot cell with a pair of slave arms. For I think that trying to perform a powder metallurgical operation would be equally difficult behind three or four feet of concrete. I must agree with you, however, that there has been no extensive production experience on sol-gel particles and that ultimately one must produce large quantities in order to be able to judge the relative merits of the processes. There are, however, a great many advantages to the sol-gel method. The main one is that it can be automated easily. It can be carried out in a system of piping and valving. That is inherently easier to do remotely, and I would think, easier to do in the open room, that is, by contact fabrication. There are some hidden expenses, though, in the sol-gel process that have to be considered. One such thing is waste water. Large volumes of waste water are generated in the use of an aqueous processing scheme, and the disposal or clean-up has to be included in the cost.

Dr. CAPRIOGLIO : May I ask Oak Ridge if there is any comment on this ?

Mr. DOUGLAS *(Oak Ridge)* : I think that it is only fair to state, at the present stage of development, one has to concede that the powder metallurgy route is economically superior to sol-gel. I would also like to comment that there is inevitably in a meeting of this kind a language barrier; thus, when one uses a certain phrase it does not mean the same in one country as in another. The sol-gel process, as we know it and practice it in the United States, is not the sol-gel process that is used in the Euratom countries. Therefore, we are comparing apples and oranges and the assessment that I would make is somewhat different from an assessment that Mr. Huddle would make as to the flexibility and the cost involved in using sol-gel. I would comment that we have considerable experience in the development of powder metallurgy processes, dating back some six years; our work in making spherical particles via sol-gel has perhaps only two years of development. Therefore, there is a time problem which hampers a fair comparison of the attributes of each process. We think that the potential of sol-gel is worth pursuing. We will establish a prototype fabrication line in which we will commence with sols and take the particles through the actual coating process in a completely integrated system. When we have had an opportunity to operate this line for an extended period, we may be better prepared to answer some of the questions as to irrecoverable losses and other economic

factors. In the Oak Ridge National Laboratory, we do not have a problem with waste water.

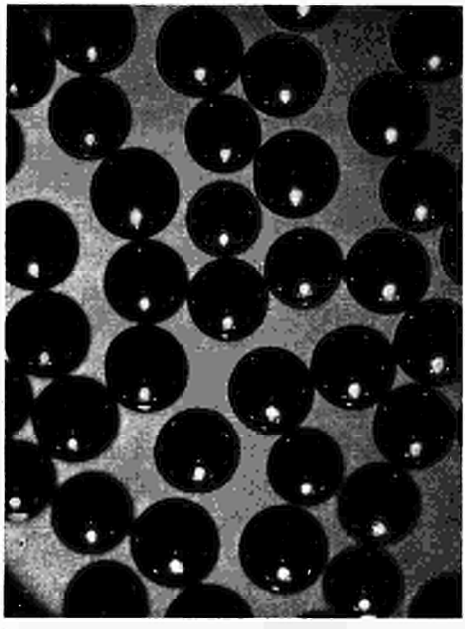
There is another question that Mr. Tytgat raised in his presentation, which concerns the size of the particles, a point Dr. Wirths also mentioned. At the present time, I would say that in using our particular sol-gel method, a maximum size that we would entertain in any economical production is perhaps 500 microns. It is true that we have made larger particles but it is not at the present time very efficient. Overall, we think that the production rate in particles of 400 or 500-micron size in one device can be carried out at a rate of about 2 kg per hour. We cannot now achieve this rate for particles much above 500 microns. However, since we make fully dense particles, our production in terms of kilograms per hour is higher than the internal gelation technique which permits one to obtain the large particles.

Dr. WIRTHS *(Nukem)* : I would like to say that we pretty well go along with Dr. Ragone. If shielding is necessary the sol-gel process should be simpler because you have a system of pipes and valves and so on. Of course, nobody of us at Nukem has thought the waste solution problem through and we do not know what impact will come from that factor. We know that in working with the sol-gel process you have less dust problems and the yield should be — from this point — better. Then, of course, one impact is from criticality. If you have to produce really large quantities, you are forced to work with a geometrically safe layout and that is somewhat more troublesome than to handle the powder. I think that one of the advantages of the sol-gel process is its capacity to produce uniform results and constant yield, while powder metallurgy yield varies.

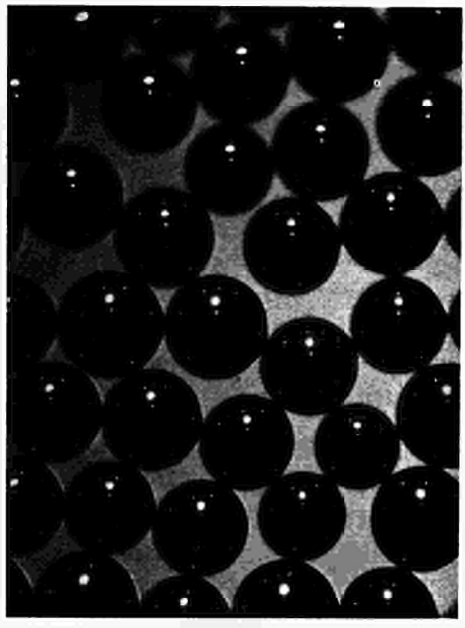
The process which we have established at Nukem is only in minor details different from those already well-known concerning the size of the sol-gel particles, however, it appears that with minor variations dense sol-gel particles up to 1.2 mm can be produced with good yields and no trouble due to cracking on heating.

Dr. CAPRIOGLIO : I am wondering if there is any other comment on this general point of sol-gel methods versus powder metallurgical methods. Has anybody had any experience in using plutonium with any of these methods ?

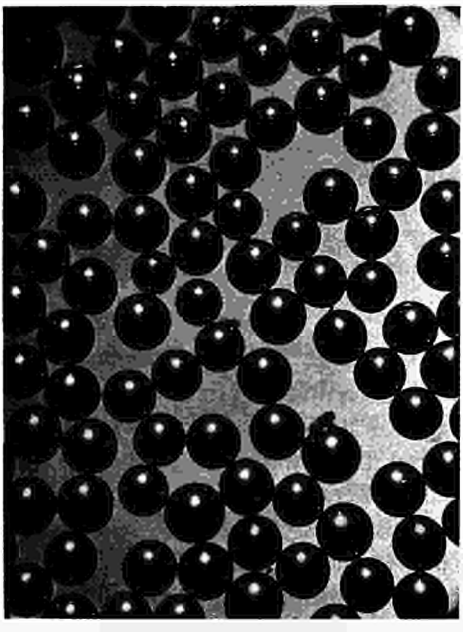
Mr. NICHOLSON *(Oak Ridge)* : At the Oak Ridge National Laboratory, we have had some laboratory-scale experience in the preparation of plutonium dioxide and mixed plutonium/thorium/dioxide particles by the sol-gel route. This process is in a very preliminary stage of development, and it is in the process of being scaled up. We cannot say that we have all the final answers for the process conditions. The first slide, is a picture of dried sol-gel microspheres. The lighter microspheres are 50 % thorium/50 % plutonium and the darker ones are pure plutonium. These spheres were formed by a process which involves precipitation of the hydroxide, peptization, droplet formation, water removal and then firing. The second slide shows the same microspheres after firing at 1,150° C; again, the dark microspheres are pure plutonium and the light brown microspheres are 50 % plutonium/50 %



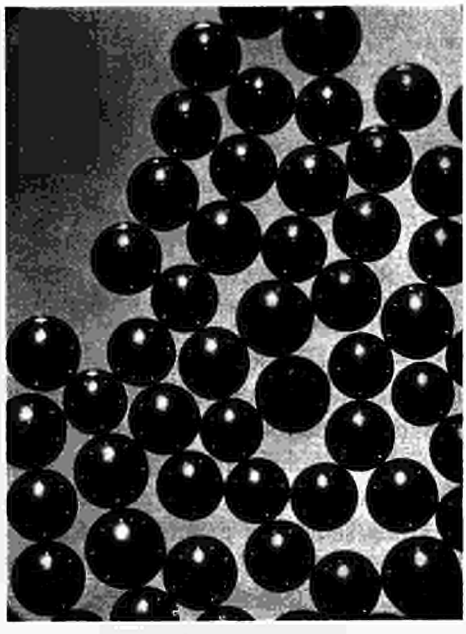
Sol-gel — 100 % PuO_2
gel microspheres
(dried — not calcined)



Sol-gel — (50 % Pu-50 % Th)O_2
gel microspheres
(dried but not calcined)



Sol-gel — 100 % PuO_2 microspheres
$\varnothing = 130 \pm 15\ \mu$ ρ » 95 % T. D.
calcined 1150° C in air



Sol-gel — (50 % Pu-50 % Th)O_2 microspheres
$\varnothing = 200 \pm 20\ \mu$ — ρ » 95 % T. D.
calcined 1150° C in air

thorium. The plutonium microspheres have a diameter of about 130 microns, and the mixed plutonium/thorium microspheres have a diameter of about 200 microns. I believe that is about all, except to mention that we have what is called a simplified sol-gel process for preparing targets for the high-flux isotopes reactor, and transuranium programme. This process consists simply of precipitating plutonium hydroxide, washing the precipitate and then firing it. The result is a very dense material which is then crushed and sized and pressed with aluminium powder for the target rods.

I neglected to mention that the density of these microspheres we have prepared is estimated to be much greater than 95 % of the estimated theoretical density of the two materials.

Dr. CAPRIOGLIO : Since I suppose that there is the intention of irradiating these materials, could I ask what the time schedule is ?

Mr. DOUGLAS *(Oak Ridge)* : The next plutonium/thorium microspheres are scheduled for irradiation in approximately six months. The pure plutonium that, as Mr. Nicholson states, is made by a simplified process, has already been irradiated for some two years. Several capsules have been examined. We are quite pleased with the results; no gross swelling or any other defect in performance has been observed.

Dr. SHEPHERD *(Dragon Project)* : I would just like to mention, since I am sure our contractors of Mol would wish me to do so, that we have done a fair amount of preliminary work on sol-gel with plutonium, and our first specimens of this will be irradiated in the next Studsvik loop; so Dragon also has some experience in this direction, but since it is such an early stage, we prefer not to discuss it, because we do not feel we could contribute anything technically useful at this stage. It is certainly going on and looking quite promising.

4. — COATING.

Dr. CAPRIOGLIO : Is there any other comment on this point? No, well, I think that time is going on, and we still have many important questions to discuss. I would suggest that we take the problem of coating, and here I think the major issue is *silicon carbide or not silicon carbide.* May I start again with Dragon on this point ?

Mr. HUDDLE *(Dragon Project)* : The initial factor that led us to develop silicon carbide coatings was a theoretical consideration of diffusion in the solid state. If you consider the structural factors that control diffusion, one comes to the conclusion that graphite in itself, particularly parallel with the close-packed planes, is a very poor material to restrict diffusion, and as silicon carbide has an almost ideal atomic structure for this purpose, it seemed the right sort of thing to develop. Since our friends at the RAE Farnborough had considerable experience in producing silicon

carbide coatings, we tried them with most encouraging results. From our experience, one of the most important aspects of silicon carbide is that it enables one to make a good coating relatively easily. Our experience of the irradiation behaviour of coated particles suggests that it is not, as one first imagines, the diffusion of fission products through the coating, that is responsable for the observed fission product emission, but the emission from :

1) broken particles, broken during fabrication of the compact;
2) contamination on the surface.

Silicon carbide enables us to make particles that, apart from the diffusion aspect, have a relatively low contamination on the surface, and in addition the particles are much stronger, and therefore less liable to breakage during fabrication. Another factor that I fear, is that the diffusion of solid fission products through the graphite structure may, in say five years, seriously impair the ability of the plain carbon coating to prevent subsequent diffusion. For this reason, particularly since it is easy to deposit, even though it costs slightly more, we in Dragon feel biased towards silicon carbide coatings, as the important thing at this stage is to have the best possible particle. I think, there is a general feeling in Dragon that in the controversy between silicon carbide versus plain pyro-carbon, we are likely to get a better return on our money by concentrating on silicon carbide. This does not mean that we are neglecting work on plain carbon coatings.

Dr. RAGONE *(General Atomic)* : On this question of diffusion in the pyrolytic carbon, Mr. Huddle is entirely correct when he says, that in parallel to the planes, this is a serious problem. Of course, we feel that by studying some of the properties of pyrolytic coatings, we should be able to put on a kind of pyrolytic carbon that will have its planes, perpendicular to the diffusion flow, so that we would not be concerned with the diffusion parallel to the planes but the diffusion perpendicular to the planes.

One more comment on this question of the degradation of the pyrolytic carbon coatings by the metallic fission products over long irradiation : of course, we have no data on that. But similarly, there is also the question of the degradation of the silicon carbide coatings over the very long burn-up periods — I do not think there is any data on that either. In both cases we will simply have to run the fuel elements for a while and see where the fission products go.

Dr. CAPRIOGLIO : I think that this problem in fact is really a problem of experience rather than a problem of decision nowadays. I would anyhow like to ask Dr. Eatherly for further comments on this.

Dr. EATHERLY *(Union Carbide)* : I might make a few comments that have some relevance to this problem. Firstly, in the fabrication studies that we have made on fuel particles, I think, as some other people also have found, we can correlate very closely the leachable uranium with the fission product release. By following the amount of leachable uranium through the process, there is no question in our

minds that the difference between 10^{-6} and 10^{-7} release rates on particles, and the 10^{-4} release rates that we see on fuel elements occurs in the moulding operations. The moulding pressures that we have used, no matter how slight, seem to damage the particles, and we can only conjecture that this is due to the angular graphite matrix materials exerting pressure on the particules during the moulding.

One of the other things that we have observed is that as we get to smoother and smoother coatings, the tendency to get leachable uranium decreases. This of course is part of the problem of orientation, diffusion and all the other things that are related to the crystal structure of the graphite coating.

The other comment I would like to make is that on straight pyrolytic materials, not with regard to fuel particles but massive bodies, a tremendous amount of work has been done with additives in order to improve the structure of the graphite. For example, materials like boron, silicon, added in trace amounts, will enhance the crystallinity of the pyrolytic immeasurably. I think one of the areas that remains unexplored in this problem, in which I must confess we have done very little, is the use of such additives rather than the use of very high temperatures in order to get smooth, highly crystalline coatings.

Dr. DE NORDWALL *(Harwell AERE)* : I would like to make a few comments about silicon carbide in view of its performance in the Pluto loop experiment. The feeling that I have about silicon carbide is that its major value is not so much for the retention of fission products which it does assist with, but its prevention of the migration of uranium, both during manufacture and afterwards. I think that in the long term, this might turn out to be more important than the retention of a fission product like barium for which it was originally used, because the retention of a short-lived fission product like barium-140 seems to be quite adequately carried out either by the fuel-tube graphite or fuel-free zone in a ball, and to this extent I would agree with the observations of Dr. Ragone.

The other point about its strength which was made by Mr. Huddle is obviously a valid one, and I think this should be borne in mind when one is considering the coating of very large diameter particles where the strength of the coating might become more important than in the smaller diameter materials.

Finally, the actual performance of silicon carbide coatings in a large experiment does not seem to be as good as one would have found from laboratory experiments and from the post-irradiation analysis of both Dragon's HPD and Studsvik experiments. One does seem to be left with the feeling that, in comparing a silicon carbide fuel with a carbon coated fuel, silicon carbide did not perform, as regards its release of strontium, as well as laboratory experiments would have lead one to suggest. Instead of an improvement of perhaps three orders of magnitude in release, one gets an improvement of about one order of magnitude only, and it does suggest, at least in these earlier silicon carbide coatings, there was a possibility of having cracked silicon carbide without having a crack which went right through. As regards the coating of oxide particles with silicon carbide, I think it is important — and I

should be glad to have information as to the relative release of metallic fission products (like barium and strontium) from oxide systems, compared with the corresponding carbide systems.

And finally, I would like to add a comment about the failed particles in an oxide system. First of all, we found in the Pluto loop that when we made a fuel element — or rather Dragon made a fuel element for us — deliberately designed to fail, it took fifteen days to do so; before the gas release reached an equilibrium value. I think this puts into perspective this question of very rapid failure concerning the point which was made, I think by Dr. Ragone, about release of fission products being more important than the release of oxygen, I would go one stage further and suggest that the fission product, that would shut a direct cooled fuel reactor down, would be iodine-131 long before either the gaseous fission products, i.e. krypton-88 or the oxygen content.

Dr. CAPRIOGLIO : I think this is a very valuable contribution to our discussion. I would ask Dr. Wirths to speak, but I would like to ask him to mention only this problem of silicon carbide since we would like him to tell us also something about the comparison between the fluidized bed and his own experience with the rotating drum device.

Dr. WIRTHS *(Nukem)* : I have a very short remark to this. As a producer, one would prefer to work with pyrolytic carbon; however, nobody knows the behaviour of pyrolytic carbon during the intended operating period for the fuel elements in the reactor, and that is the reason why we are happy that in the Dragon project, a second type of coating will be developed. We hope that one day we will have two good coatings.

Dr. CAPRIOGLIO : Is there any further comment on this question of silicon carbide ?

Mr. SHENNAN *(UKAEA, Springfield)* : I would like to confirm the views of previous speakers on the rôle of silicon carbide coatings in reducing the level of uranium contamination and increasing the strength of the coated particle. However, we have found uranium contamination of pyrocarbon coatings to be largely a function of the concentration of uranium in the leach liquor. Thus, the contamination levels may be reduced by removing the major source of this soluble uranium, which are sooty deposits adhering to the batch, before leaching. We find, for example, that coating at 1,500° C and removing deposits by acetone washing followed by leaching in nitric acid produces coated particles with the low contamination levels of silicon carbide coated particles. With respect to the strength of coatings, we find that fuel compacts containing silicon carbide coated particles may be extruded without significant failure of the coats, but this is not true for pyrocarbon coated particles.

Mr. FORBES-GOWER *(CEGB, London)* : I wonder if, Mr. Chairman, I might make a comment as an operator. It is in verification of what Dr. Shepherd said when he

thought that the fabrication cost was a small matter compared with the reliability of the fuel. I should like to point out that the situation in Great Britain is different from that in America, relative to security of supply. The one thing that is more important to us than any minor difference in fuel cost due to fabrication, is the cost of outage time; the large 1,000 MW reactors of the type of which we are talking, would cost us about eighty thousand pounds a day if their outage were to be increased. In this respect I notice that Mr. Valette was saying that the present experience was that the carbide fuel was showing less failure than the oxide and therefore, although I am not in any position to comment on carbide or oxide, there seems to be a very strong argument as an operator, in favour of silicon carbide coatings, and of carbide fuel versus oxide fuel.

Dr. CAPRIOGLIO : Since this is the voice of the client, I am sure everybody will listen to it. Is there any further comment ?

Dr. ROEMBERG *(Dragon Project)* : From the information so far, it seems that the quality of the particles is linked very closely to the R/B ratio as found under ideal and perhaps slightly unrealistic gas conditions. In a corrosive atmosphere, obviously the silicon carbide coated particles must be superior. I am, therefore, wondering if it would not be beneficial, if some of the quality tests could be carried out under slightly more realistic gas conditions.

Mr. TRAUGER *(ORNL)* : I might comment that we plan to do materials compatibility studies on irradiated particles in the near future; we will be conducting a number of tests with atmospheres of helium containing water vapour and other impurities.

Dr. SHEPHERD *(Dragon Project)* : Just a very brief remark, in case anyone has gained the impression that Dragon is walking on one side of the fence and everybody else on the other. I would just remind you what has been said already : that Dragon is using both oxide and carbide particles, using both sol-gel and powder metallurgy, straigth pyro-carbon as well as silicon carbide, and furthermore we are going to be irradiating the fuels at much higher temperatures than we believe are acceptable, and also using plutonium as well as uranium-235.

Dr. CAPRIOGLIO : I do not think, there was any doubt in anybody's mind that the discussion was really mainly on preferences rather than on programmes. After all, the differences are so marginal, at such an early stage of development, that it is very hard to make clear-cut decisions nowadays. Any further comment on the silicon carbide coatings ?

Dr. RAGONE *(General Atomic)*: I think that we should in this session pose a question for the reprocessing people in the next session. The question concerns the reprocessing of silicon carbide coated particles. How much more difficult than the reprocessing of simple pyro-carbon coated particles will it be? To add to Dr. Shepherd's comments, everyone in this business is hedging his bets. No one feels

so certain of his stand that he is willing to perform just one set of experiments. I have a brief comment on production costs. When we mention cost reduction we, of course mean cost reductions consistent with reliable and safe operation.

Dr. CAPRIOGLIO : I think before leaving this subject of coating, we should ask Nukem to tell us something about their experience in the comparison between the fluidized bed and the rotating drum on coating.

Dr. HACKSTEIN *(Nukem)*: Yes, we have only little experience comparing rotating furnace and fluidized bed because we work just at this time on a ten-gram-scale with our rotating furnace. But it looks promising, the α-count is very good and the xenon release compared with those particles coming out of the fluidized bed are better just at this moment. The economical is open, but we start to build a rotating furnace of about one-kilogram-scale and then we could say more on this.

Mr. TYTGAT *(Euratom)* : From recent discussions with Nukem I learned that they have developed a rather interesting technique, using a fluidized bed, which might present rather new economic advantages.

Dr. HACKSTEIN *(Nukem)* : We have done in the last few months some work on coating under vacuum, under a contract of the German Ministry. We worked with a fluidized bed on a 1-kilogram-scale and a range of about 30 to 300 torr. In this range, we get all structures we want, either laminar and columnar, fine columnar and rough columnar, and we need only 5 to 10 % of the argon or argon + methane, used when working under normal pressure. In this case, we spare a lot of argon, we can work with a composition up to 90 % methane and we have no soot. The particles are very good. The advantage of this method is that we spare argon and we have better particles, that means the gas content in the coatings is smaller than in the ones of particles worked under normal pressure. The crushing strength (if you compare equal structures and equal layer thickness, under normal pressure and under vacuum) is better when we work under vacuum; the alpha count is much better. The deposition rate is between 4 to 20 or 30 microns per hour. Further work is going on.

Mr. DE BACCI *(Euratom)* : I think it has been said during this session regarding coating and kernels, that we have to make the best particles that we possibly can. I think that this assumes that we want to design a fuel element which allows a burn-up as high as possible. This implies that we are designing an advanced converter. Without anticipating too much what will be said tomorrow in one of the sessions, we shall see that if we want to design a breeder based on the HTGR concept, we have to restrict the burn-ups to, say, about 30,000 MW/day per ton. In this case, we cannot afford to use a fuel element which is the best that we possibly can, we just have to make it as good as it is needed, and we might dispense with silicon carbide coatings and perhaps with carbide kernels.

Dr. CAPRIOGLIO : Is there any other comment in particular or first thoughts about the Nukem contribution, on their coating under reduced pressure ?

Dr. RAGONE *(General Atomic)* : Just a question concerning an analysis of the hazards during the operation of the vacuum coater. Operating under a vacuum or under reduced pressure brings about the problem of the leakage of water or air into the reaction chamber. There is hydrogen in the furnace. If a crack developed in the furnace you bring oxygen in and possibly get explosive mixtures. What kind of considerations have been given to the safety of this kind of operation ?

Dr. WIRTHS *(Nukem)* : We are not afraid that this will happen. An explosion cannot happen since you have almost no pressure in the system. If you have only 30 or 50 torr of methane you can admit air. Of course, the design of the furnace has to be done so that you can really apply vacuum. As about the possibility of water getting into the furnace, that also depends on the design. What would happen in your case depends on the design, whether water can come into the furnace at all.

Dr. CAPRIOGLIO : Thank you very much. On this matter of coating, Mr. Tytgat and myself think that there is one question that is of great importance, namely, what is the *maximum size of fluidized bed* that is likely to be built in terms of criticality, hazards, and technical feasibility. Does anyone wish to make a comment on this ?

Dr. RAGONE *(General Atomic)* : I can make some comments on this. Of course the attractiveness of having larger sized beds is obvious. They are cheaper to operate. The extent to which one goes depends on criticality safety rules. If one is limited by the amount of uranium that one can put into a fluid bed, that is if one has a mass limit on criticality, there is not too much to be gained by going to greater diameters. We tend more to use geometrical limitations, and have been able to coat in as large a bed as 5 inches in diameter. We have coated batches, as high as 7 kg. There are considerable economic advantages to do so.

Mr. HUDDLE *(Dragon Project)* : Could I just add one small point which may be important? If you have a very large bed, and something goes wrong then you have a major reject problem. When one is scaling up, this is a point that should be borne in mind.

Dr. EATHERLY *(Union Carbide)* : I will come in on that one. Our experience has been very sharply that the larger the batch size we use, of course the more we lengthen the time that the particles are in the furnace, and thereby the better control we have over the particle bed. Within reason the larger beds are more stable, the particles are rotated more uniformly, agitated more uniformly throughout the entire volume of the bed, the temperature variations can be kept under better control and averaged out better. So that our experience has been that reliability increases as our batch size increases.

Mr. TAYLOR *(UKAEA, Harwell)* : On the question of scale-up, I would like to raise the topic of bed height. Is there any information available on the effect of bed height on the efficiency of coating, with special respect to the amount of soot

formation? And on this same aspect, in the work carried out on rotating drum contactors, and vibrated beds, where the gas is presumably introduced above the bed level, does the soot formation become much more serious in these types of apparatus ?

Dr. CAPRIOGLIO : Any answer to this question? Apparently, there is no answer for the moment.

5. — FUEL ELEMENTS COSTS.

Any further comments on this question of coating? If not, I think that we have now some minutes left to have an exchange of views on the cost of making these fuel elements. The papers that we have received seem to be quite different from each other. Since a large experience is not yet available to anybody, it is only natural that it is very unlikely to arrive at similar assessments. One of the questions that has been raised in detail was this question of *losses*, and I understand that there is a rather serious difference between the assessment of Dragon and Oak Ridge : Is it just a difference of being more or less conservative, or is there likely to be any serious intrinsic difference of evaluation ?

Dr. RAGONE *(General Atomic)*: I think, one cannot criticize Mr. Huddle's paper for quoting 1 % as irrecoverable loss. However, when you look at the fraction of total cost involved (see table 5) Dragon paper p. 13, 10 % of the total cost of coated particle production is involved in these losses, so it is an important thing to discuss. We feel that we can operate at much less than that; and I think that our Peach Bottom experience was less than 1/2 % loss. But in order to get reliable loss figures one has to run many thousands of kg and there has not been too much production in the world of these materials. I would enter the figure of less than 1/2 % loss as my best guess.

Mr. HOUDAILLE *(Dragon Project)* : I quite agree with Mr. Ragone that this question of loss is rather a conservative figure. But in fact, we took it because we saw that this was the only realistic value we could put in our estimate. The fact that this 1 % loss gave 10 % of the total fabrication cost is just a very good incentive for us to chase the losses and try to reduce them by any means.

Mr. LOTTS *(ORNL)* : We have some experience which is on another process but with the fabrication of U^{233}/thorium. This pertains to the Kilorod facility and its operation at Oak Ridge, where we fabricated approximately one thousand kilograms of 3 % weight U^{233}/97w/o thorium.

In all of this operation, which involved powder techniques, that is crushing, grinding — similar operations that we are speaking of here — we had 0.1 % irrecoverable loss. I think another point in regard to this is the differences in apparent losses and actual losses, apparent losses being due probably to the inaccuracies of the chemical analyses.

Dr. WIRTHS *(Nukem)* : For several years now we are handling 90 % enriched uranium in reasonable quantities for MTR fuel elements. Experience has shown that with time losses can be reduced without special efforts. Furthermore, you can avoid almost any loss if you are ready to spend money. So after some time, it is a simple matter of calculation and not a matter of principle.

Dr. CAPRIOGLIO : This seems to me the voice of common sense, and I am wondering if this is not the statement at which to stop this discussion on losses. There is one thing on this cost problem that I have been myself very much interested in, to see that everybody is being rather conservative about the *size of the industry.* People talk about 1,000 MW or 4,000 MW. In fact if a reactor system is to be considered economically successful, it has to be built for much larger capacities than this. Any economic assessment of the fuel elements should be based as well on a very large production; the system itself will have been an economic failure, if it was only going to be built on total capacities of a few thousand MW. One has only to think of the amount of money that has gone in research and development, for instance, all over the world. If one wants to amortise this money over a few thousand MW, one would find it very difficult to produce electrical energy at a reasonable cost. So, also for the fuel element side, I would like to stress that our assumptions have been rather conservative on the size of the industry.

Mr. LOTTS *(ORNL)* : I think that what I will say is contained also in the paper this afternoon. We found that, particularly in the assembly operations, it is necessary, in the case of HTGR, to get to fairly large capacity before you have complete utilisation of your equipment. So therefore, you would expect the cost to be reduced at least up to, say, one ton a day, which is a substantial amount of fuel. The other point I would like to make is that certainly in order to realize the advantages of mass production, it is necessary that we have standardization of the fuel configuration, particularly that part which pertains the assembly. You very fast reach the maximum material that you can handle in one coater or in one conversion apparatus, but not so with the assembly operations. So if one had standardization of that phase of the fabrication, he could realize further saving at half capacity.

Dr. WIRTHS *(Nukem)* : We agree with Mr. Lotts that we are extremely far away from the point where we can say that we are producing fuel economically. First let me say that we have not used our capacity by working in three shifts, which would be very economical. The second point is that we can scale up apparatus.

I do not dare to give any figure what this would mean, scaled-up apparatus and works in three shifts, but I think it is not too different from what Mr. Lotts said. And still afterwards, of course, we could lower production costs, if there is one line beneath the other. We are far away from this, we work on one line essentially. So we are really very far from economical fuel production.

Dr. ROEMBERG *(Dragon Project)* : If we envisage increasing the size of the production plant, have we not to bear in mind that this adds to the *transport problems* which have then to be balanced against the gains of increasing the size of production?

Dr. CAPRIOGLIO : I personally feel that it does not cost too much to transport fuel elements as compared to what it costs to make them for the moment. Any further comment?

Dr. WIRTHS *(Nukem)* : In our experience, there is only one transport problem, which is to get your UF_6 from Oak Ridge to Europe; that is rather expensive. But the transport within the factory does not play any rôle.

Dr. RAGONE *(General Atomic)* : I have one very specific question on the paper by Huddle and Houdaille and *al.* on this question of *conversion of* UF_6 *to* UO_2; that particular cost amounts to 21 % of the total coated particle production cost, and would certainly seem to be one that would come down sharply with rising volume.

Mr. HUDDLE *(Dragon Project)* : We checked this cost with our collaborators in Europe and also with friends in the United States, and the cost we have used, is that in Europe. We know the cost in the United States is approximately half our own value, but in Europe that price is doubled due to transport, etc. The figure, we have used, is the cost we think one would have to pay here in Europe, if the programme was started now.

Mr. VALETTE *(Euratom/THTR)* : I should like to ask for some comments about the *cost of graphite.* The figure quoted in the Dragon report is 600 pounds sterling per ton of machined graphite. Taking the figures which I believe are available at THTR, a machined ball costs approximately 1 pound sterling (appr. DM 10,—). One ball weighs approximately 200 grams, so it makes 5 pounds sterling per kilo, makes 5,000 pounds sterling per ton. So there is a factor of 10 in cost of graphite. I would like to have the comment of someone.

Mr. HOUDAILLE *(Dragon Project)* : Obviously, now it may be a factor of 10 in cost of graphite. We hope that by the time we build a reactor of about 2,000 MWe, the cost of graphite will be reduced. We felt, in writing this paper, that it was not so obvious that we needed actually very much elaborated graphite. We could probably go to a cheaper brand, and we were thinking of using the kind of graphite, being developed in England for instance, for the AGR reactor, which turns out to be this price of 600 pounds sterling per ton including machining. Moreover, we stated in our paper that this price refers to the weight of the extruded graphite prior to any machining and not to the weight of the finished structural block.

Mr. HUDDLE *(Dragon Project)*: Could I just add a word. For the purposes of this paper, we took an actual possible design and calculated how much could be cut from an extruded and graphitized block. The price of the improved graphite was estimated knowing that the cost of high-grade graphite is about 375 pounds sterling a ton. We feel therefore that 600 pounds sterling a ton was a realistic figure. It does not include the cost of graphite in the fuel compact, but just the structural graphite used in the core.

Dr. CAPRIOGLIO : There is one question that seems important to me, which is the *use of free particles in a fuel element.* It is obvious that the possibility of using free particles would be a serious advantage on economic grounds, but is there any experience so far or is this problem being thought over by all the interested people in high-temperature fuel ?

Mr. HOSEGOOD *(Dragon Project)* : I have a question to ask about the use of free particles in fuel elements. They have to be confined in some form of graphite container. I would like to know, if it is considered the correct practice to put a hole in the graphite container so that gas can pass freely in and out, or if it is intended to make it pressure tight and sufficiently strong to withstand the pressure variations that it will encounter in service.

Dr. RAGONE *(General Atomic)* : We have, of course, been interested in lower R/B in loose coated particles, and mainly there are several advantages one can see in terms of manufacturing cost; it is easier to manufacture one of these; and secondly it has considerable advantages for reprocessing, one would be far better off, if one could simply pour loose coated particles into the reprocessing scheme rather than the whole fuel element. These are clearly the advantages that one hopes for. We are currently running an element with loose coated particles in our loop, and have had no difficulty with this at all, that is, thermal conductivity of the bed does not seem to influence the operation in any way. It is true that a loose bed has a lower thermal conductivity than one with a matrix, but with proper design, that is making the minimum dimension for heat transfer a little lower, you can take this into account and operate successfully. To answer the question on pressure variations, we consider the graphite strong enough to take a pressure variation. Of course, you recognize that it is not an entirely tight container, the permeability of the graphite that would be used, would be on the order of about 1 sq.cm. per second, and so pressure variations could not last for very long. The attitude that we have however is this : it is attractive to use loose coated particles; however, this is a question of operation and safety. We are going to do more and more experiments on loose coated particles. If they can be used, we will use them, but again consistent with smooth operation of the reactor.

Dr. SHEPHERD *(Dragon Project)*: I would question the assumption that it would be cheaper to work with loose particles. Loose particles have to be contained in something, which means that you have to machine some sort of box. In prismatic fuel, I believe, that would be more expensive than the process which we envisage of compacting overcoated particles. It may be that one of these alternatives is technically superior to the other and that might be the deciding factor. But I certainly do not think, there is any grounds for saying that loose particles will be cheaper than compacted particles.

Mr. VALETTE *(Euratom/THTR)* : I should like to ask Dr. Eatherly who, I think, mentioned that the matrix of the irradiated fuel sphere has not shrunk during irradiation : in which fast flux was this matrix irradiated ?

Mr. TRAUGER *(ORNL)* : The irradiation to which Dr. Eatherly referred was done in the ORR pool-side facility, which has a rather low fast flux. I do not recall the exact exposure for that element, but it probably was around 10^{20} NVT. We have irradiated other matrix elements as mentioned in the paper, up to about 2×10^{21} NVT. We found about 1 % shrinkage on the machined shells, and about 2 % on molded spheres.

Prof. SCHULTEN *(KFA/THTR)* : I should mention that in our development we have also made some irradiation experiments with loose coated particles for the ring annular type. The experience which we got up to now with Dragon particles is quite good, and also in our case, we do not believe that the problem of changing the pressure is really a problem, because the shell is strong enough to avoid these problems.

Dr. CAPRIOGLIO : I think that we have done a good job this morning and that we deserve a bit of rest.

STUDIES ON THE REPROCESSING AND REFABRICATION OF HTGR FUELS IN THE OECD DRAGON PROJECT (1)

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ABSTRACT

The aims of the Dragon Project fuel reprocessing studies are broadly outlined. Preliminary results of assessment and experimental studies on the head-end treatment of spent fuel elements commensurate with the centralised reprocessing requirements of an integrated HTGR power system of 10,000 MW(th) are presented.

Proposals for the decontamination of irradiated Th/U fuels by solvent extraction at the rate of 60-70 kg (Th/U) per day to produce either uranyl nitrate solution for rapid fuel fabrication in a lightly shielded facility or thorium/uranyl nitrate solution for conversion to fuel in a heavily shielded facility, are presented and discussed.

Relevant results of experimental studies on the preparation of particulate oxide and carbide fuels by powder metallurgical and sol-gel techniques are presented and schemes for the refabrication of such fuels from high enrichment uranyl nitrate and uranyl/thorium nitrate solutions are outlined.

1. — INTRODUCTION

1.1. — THE DRAGON PROJECT FUEL REPROCESSING ASSESSMENT STUDY.

Fuel cycle optimisation studies previously made by Dragon Project have demonstrated the significance of the contribution of reprocessing and refabrication costs to the unit cost of electricity produced from power reactors utilising the uranium-thorium cycle [1]. Although Lane [2] has made fuel cost estimates for various thorium breeders and converters it will be several years after the thorium cycle is adopted before fuel reprocessing can be carried out on the scale required to furnish significant fuel cost data. For this reason the Dragon Project decided early in 1963 to initiate an assessment study of the feasibility and costs of reprocessing and refabricating uranium-thorium carbide fuel. It was appreciated that accurate predictions could not be expected from such a study but it was considered that cost estimates would be obtained which could be used in optimisation studies with greater confidence than those available hitherto.

For the purpose of the study it was assumed that a central reprocessing/refabrication plant would be required to serve a programme of 10,000 MW (th) spread

(1) Dragon Project Report 338, April 1965.

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over several reactors using highly enriched uranium-thorium dicarbide fuel. Based on the conclusions of the early burn-up optimisation studies it was further assumed that the fuel would have a fifa value of 1.6 (burn-up of 150,000 MWd/(t) and would remain in the reactor for four years. An annual reloading cycle of one quarter of each reactor core was specified (300 days per year at nominal power rating) and an average core rating of 1.2 MW(th)/kg of initial fissile metal was postulated. From the fuel cycle calculations an "N"-value (thorium/fissile uranium atomic ratio) of 12 and an "S"-value (total carbon/fissile uranium atomic ratio) of 3,000 were specified for the refabricated fuel.

It was decided that the fuel should be of the fission product-retaining type, comprising kernels of average composition (U, Th)$C_{2.4}$ coated with a triplex layer of pyrolytic carbon-silicon carbide-pyrolytic carbon, and pressed into compacts with graphite powder. Whilst specification of the fuel element geometry could be only arbitrary, inasmuch as fuel development and experience of operation of HTR's such as the Dragon Reactor Experiment would make any reference geometry obsolete, it was necessary to adopt a geometry for purposes of assessing the type of fuel handling equipment. Hence an annular design of unpurged fuel compact in a hexagonal graphite lattice was envisaged. The reference fuel element is not described here in greater detail since the choice was, as stated, arbitrary.

J. L. Schlösser of Dragon Project has calculated the approach to an equilibrium fuel cycle composition for reference fuel, fabricated initially from thorium and 93 % enriched U^{235}. The quantities of heavy metal in the discharged fuel from the equilibrium fuel cycle of the reference cores are as shown in Table 1 (100 % utilisation factor of the reprocessing plant assumed).

TABLE 1. — Quantities of Heavy Metal in Discharged Fuel from Equilibrium Fuel Cycle for 10,000 Mw(th) Reactor Programme

Th^{232}	60 kg/day	
U^{232}	3 g/day	
U^{233}	3 kg/day	
U^{234}	1 kg/day	
U^{235}	1 kg/day	
U^{236}	2 kg/day	
U^{238}	0.5 kg/day	
Pu^{239}	20 g/day	if Pu is recycled, otherwise total Pu is less than 5 g/day.
Pu^{240}	10 g/day	
Pu^{241}	10 g/day	
Pu^{242}	10 g/day	

It was assumed that 200 days cooling would occur before reprocessing commenced, hence Pa^{233} is not included in the above although it would amount to about 10 % of the U^{233} concentration in the discharged fuel. In summary, therefore, the

daily throughput of the reprocessing plant serving a 10,000 MW(th) reactor programme would be as follows if the plant load factor were 300 days per year :

Thorium	73 kg/day
Fissile uranium	4.9 kg/day
Non-fissile uranium	4.3 kg/day
Plutonium	6-60 g/day
Graphite	1,060 kg/day
Fission products	11 kg/day

The reprocessing and refabrication routes specified and the reasons for their choice are described in Sections 2 and 4. Within the restricted terms of reference of the study it was not possible to mount the substantial development programme which would ultimately be necessary for a full engineering assessment. The philosophy adopted by the Project was that experimental investigation would be made wherever possible of key stages of reprocessing and refabrication for purposes of defining the flow sheet. In a few selected instances pilot plant studies were also planned but these were not comprehensive. The results of the experimental work which has been completed to date in support of the study are reported in Sections 2 and 4.

Contracts for the assessment study were placed by Dragon Project with the UKAEA (Engineering Group, Risley and Production Group, Windscale) for the basic head-end process, fission product decontamination and separation stages, and with CNEN, Rome, for an alternative head-end process and the refabrication stage up to the production of uncoated fuel particles. The intention was that the particle coating and fuel element production stages would be assessed within the Project from its experience in these fields.

At the date of the preparation of this paper (March, 1965) the study had proceeded to the point where the flow sheets for all stages of the process had been selected. It is estimated that the work will be completed, and order-of-cost estimates for reprocessing/refabrication available, at about the end of 1966. The present paper is, therefore, an interim report only on Dragon Project's activities in these fields.

2. — Head-End Treatment of Spent Fuel

2.1. — Possible Processes.

A variety of processes has been suggested, particularly by investigators at ORNL, [3, 4, 5] and tried out on a laboratory scale for recovering uranium and thorium from SiC coated (U, Th)C_2 kernels dispersed in graphite compacts. A brief description of these processes, the Dragon Project's reason for choosing the mechanical grinding and leaching method and a summary of the laboratory data obtained in support of the choice are given.

2.1.1. — *Combustion-dissolution Processes.*

(*a*) Combustion.

Simple combustion of the fuel at 800-1,000° C in oxygen will remove all matrix carbon leaving the SiC coated particles intact. (SiC is only attacked rapidly by oxygen at much higher tempeature : in the range 1,800-2,000° C.)

The method could be used to reduce the volume of fuel material that has to be subsequently reprocessed.

There may be, however, severe containment and handling problems associated with the large volume of active exhaust gases and volatile fission products emanating from broken fuel particles : if combustion were adopted $\sim$ 2,000 m^3 of active off-gas per day would be produced in the reference reprocessing plant.

Combustion after grinding would produce an extremely active exhaust gas stream but would leave an oxide residue of uranium and thorium soluble in 13 M HNO_3/0.04 M HF.

(*b*) Pyrohydrolysis.

This method which is a variant of combustion is being developed at ORNL where it has been found that irradiated uranium and plutonium monocarbides, when exposed to steam at atmospheric pressure at 700° C to 800° C, are converted to the corresponding oxides which can readily be dissolved in HNO_3 to yield a solution suitable for solvent extraction. In the ORNL studies the residual carbon after treatment was negligible and the gaseous reaction products were H_2, CO and CO_2. Caesium was the principal fission product volatilised and was trapped in the condensate.

For Dragon fuel this process is unattractive because the graphite would not be significantly attacked at these temperatures, hence a pretreatment such as anodic disintegration would be necessary for separation of the graphite.

2.1.2. — *Volatility Processes.*

(*a*) Fluoride.

The complete elimination of free carbon from the material to be fluorinated is imperative in order to avoid explosions initiated by fluorine-carbon compounds when processing SiC coated fuel particles. Grinding and combustion would therefore probably be perequisites of the fluorination route for removing free carbon from the kernels,

A further disadvantage of the fluoride volatility process is the presence of non-volatile fluorides which may dissolve some of the volatile UF_6 to form solid solutions. In addition, this method is unsuitable for recovery of thorium from the non-volatile fission product fluorides.

(*b*) Chloride.

Unless the SiC coated particles were fractured before chlorination, a temperature above 1,200° C would have to be employed to volatilise the coating. Furthermore

some fission product chlorides would also be volatilised with the thorium and uranium, necessitating further decontamination steps.

2.1.3. — *Grind-Leach.*

This technique, studied initially in Dragon Project by R. O. Lingjaerde [6], has been utilised successfully on a laboratory scale and is the Project's first choice of head-end process for the reference reprocessing plant. The process involves mechanical grinding of fuel compacts to rupture the fuel particles, followed by an appropriate acid leaching to dissolve the fissile-fertile material. Treatment for the destruction of organic matter and conditioning by adjustment of acidity produce a feed solution suitable for the subsequent solvent extraction of fission products.

Moreover, all the other processes which have been briefly considered above will most likely require a grinding step to penetrate the SiC coating although for the combustion route the amount of material to be crushed would be greatly reduced. Thus, because of the ease with which the laboratory scale grind-leach experiments were carried out and since the essential simplicity of the apparatus involved appeared to outweigh the advantages of reducing the volume of material to be processed by a high temperature oxidation treatment, the grind-leach head-end step has been adopted for the Dragon reprocessing assessment study.

As a potential refinement, anodic disintegration prior to grinding is being studied also on behalf of the Dragon Project by CNEN, Casaccia, Rome, as a method of separation of the graphite from SiC coated particles, to reduce the volume of material to be ground and leached.

2.2. — GRIND-LEACH EXPERIMENTS AND RESULTS IN DRAGON FUEL LABORATORIES,

2.2.1. — *The Leaching of Th/U from Ground Particles* (PyC/SiC/PyC Coated).

A series of leaching experiments has been carried out under identical conditions, constant parameters being the quantity of ground particles (2 g) and the volume of acid (100 cm^3). The variables studied were leaching time, acid strength and particle size of the powder.

The results have shown that :

(i) It is necessary only to crack open the fuel particle coating to ensure a quantitative recovery of the kernel material by acid leaching.

(ii) Quantitative recovery of uranium and thorium is achieved by attacking the ground fuel particles with pure HNO_3 at a concentration of not less than 8 M for a minimum time of 4 h, a practical acid concentration being $\sim$ 13 M.

(iii) Nitric acid dissolves out the kernel material quantitatively even when the carbides have decomposed, i.e., by hydrolysis after exposure in a damp atmosphere. Also in this case the addition of HF as catalyst for dissolution of thorium is not necessary.

2.2.2. — *The Leaching of Th/U from Ground Compacts*

A series of experiments was carried out on PyC/SiC/PyC coated particles [(Th, U)$C_{2.4}$ kernels] dispersed in a Dragon-type graphite compact, in which the ratio U : Th : C was 1 : 8 : 100. For each experiment a sample of approximately 200 g was used and the experimental conditions which conformed to the treatment proposed in the flow sheet (see below), were as follows :

(i) *Grinding* : Size used < 251 μm and < 75 μm

(ii) *Hydrolysis* : 4 M HNO_3 : ~ 2.6 cm^3/g powder, 1 h boiling

(iii) *Leaching* : 13 M HNO_3 : ~ 1.6 cm^3/g powder, adjustment of acidity by addition of 16 M HNO_3 and of volume by distillation, 6 h boiling.

(iv) *Filtration* : The cake was washed four times with distilled water. For each washing a volume equal to the volume of the initial filtrate was used.

By comparison of the results obtained with < 251 μm and < 75 μm graphite/carbide powders it appears that the following advantages were obtained by grinding the compact to < 75 μm :

(*a*) Better uranium recovery. The U retained was reduced from 1.75 % to 0.45 %.

(*b*) Reduction of the overall operating time of the experiment (hydrolysis leaching-filtration).

As might be expected, the retention of liquor is higher in the filter cake from the finer powder but virtually all the uranium is removed in three washes. Even after the second washing more than 99 % of the total U is leached out, the operation itself being quick and simple to carry out on the laboratory scale.

A problem which requires more study are the organic soluble carbon and carbide degradation products produced by the leaching with nitric acid. Their presence could interfere with the uranium and thorium recovery during solvent extraction and it is proposed to use an oxidative degradation with permanganate to follow the leaching to minimise their interference. This aspect is being studied by CNEN, Rome.

2.3. — Proposed Flow Sheet for Basic Grind-Leach Head-End Process.

Irradiated fuel will be received in mild steel cans which have been cooled for not less than 150 days in the reactor ponds. After 50 days further cooling at the reprocessing plant, they are fed to the head-end process which comprises the following operations (Fig. 1) :

(1) A decanning operation where the fuel cans, each containing up to seven elements, are removed. The cans are crushed and baled for disposal to an active silo.

(2) Two-three stages of milling and grinding are carried out under an inert atmosphere of N_2, to produce a powder suitable for leaching. The off-gases are filtered and scrubbed with caustic soda before discharge from the stack.

(3) The powder, by weight control, is conveyed to the dissolvers where the carbides undergo a preliminary decomposition in 4 M HNO_3 followed by a leaching in 13 M HNO_3. For these operations HNO_3 recovered from the subsequent stages is used with a make-up of about 50 % fresh HNO_3. An oxygen feed is

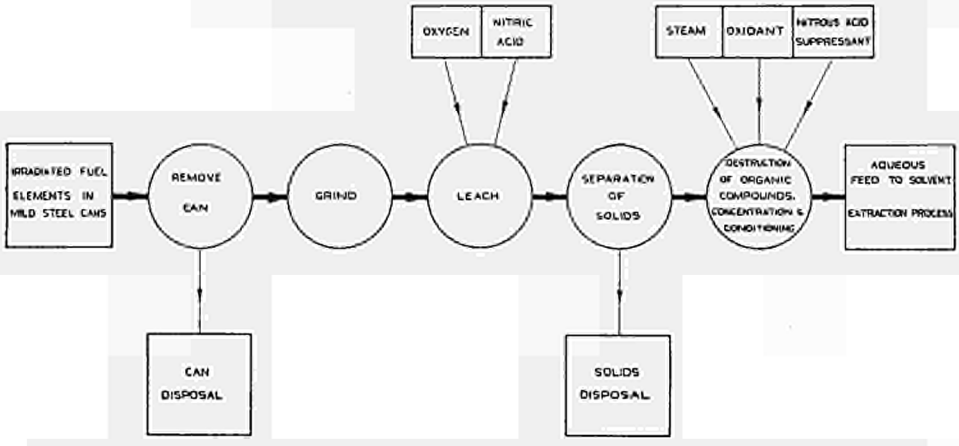

FIG. 1. — Block Diagram : Head-end Section of Reprocessing System using the Grind-leach Process.

provided to oxidise NO evolved during the hydrolysis to NO_2 to facilitate removal of the nitrous gases before discharge to atmosphere.

(4) The leached suspension is pumped out from the dissolvers, the solids are removed by filtration and after washing with water are disposed of.

(5) The solution is then concentrated and refluxed with potassium permanganate, which promotes degradation of the organic soluble material. The solid residue of MnO_2 is removed by centrifugation and washed with water.

6) The resulting solution is first concentrated and then steam stripped to an acid deficiency of 0.36 M.

A final feed adjustment is performed after destruction of nitrous acid with $CO(NH_2)_2$ or H_2SO_3 NH_2 to produce a solution suitable for the solvent extraction of the following composition :

~ 0.1 M acid deficient, ~ 0.13 M U, ~ 1.1 M Th.

If graphite separation by anodic disintegration is found to be technically feasible it can be inserted between the decanning and grinding stages of the head-end process. Two types of electrolysis cell are being developed by A. Moccia, CNEN — a series cell suitable for disintegrating fuel compacts after mechanical separation from the graphite element and, alternatively, a contact cell which can be used with the entire fuel element if mechanical removal of the compacts is not possible.

3. — Fission Product Decontamination and Fissile/Fertile fuel separation

Pittman of the USAEC summarised at Gatlinburg in 1962 [7] four basic routes for reprocessing and refabricating uranium-thorium fuel. The assessment study of

fuel recycling being made by the Project will estimate the costs of two of these routes which are considered to be of major interest, viz. :

(1) Intermediate ("minimum") decontamination from fission products followed by refabrication of fuel from the coextracted U,Th product in remotely operated, heavily shielded facilities.

(2) High ("maximum") decontamination followed by direct refabrication in lightly shielded equipment. It is necessary in this route to separate the thorium from the recycled uranium and to store it long enough (about 15 years) for the high γ-energy daughters of the U^{232}/Th^{228} chain to decay to an acceptable level. It is also necessary for refabrication to be completed quickly following decontami-

TABLE 2. — Fission Products in Irradiated HTGR Reference Fuel

Fission Product	Curie/kg (U + Th) in Spent Fuel after 200 days Cooling	γ-Activity curie MeV/kg (U + Th)
Kr^{85}	4.6×10^{2}	6.0×10^{-2}
Sr^{89}	2.70×10^{2}	
Sr^{90}	5.2×10^{2}	
Y^{91}	4.9×10^{2}	1.9
Zr^{95}	5.87×10^{2}	4.38×10^{2}
Nb^{95}	12.0×10^{2}	9×10^{2}
Ru^{103}	5.82×10	2.73×10
Ru^{106}	9.32×10	
Sb^{125}	1.2×10^{1}	5.5
I^{131}	4.6×10^{-5}	
Cs^{134}	3.0×10^{2}	4.5×10^{2}
Cs^{137}	4.8×10^{2}	
Ba^{137}	4.8×10	3.2×10
Ba^{140}	1.2×10^{-1}	2.2×10^{-2}
Ce^{141}	8.0×10^{1}	6
Ce^{144}	3.3×10^{3}	1.4×10^{2}
Pr^{143}	2.3×10^{-1}	
Nd^{147}	1.2×10^{-2}	2.8×10^{-3}
Pm^{147}	1.3×10^{3}	
Sm^{151}	1.2×10	7×10^{-3}
Eu^{154}	4.4×10	4.0×10
Eu^{155}	1.8×10	1.3×10
Total γ curie MeV/kg (U + Th)		2×10^{3}
Total γ curie MeV/day through reprocessing plant		1.7×10^{5}
Decay heat per element on receipt at reprocessing plant, W. .		210

nation and separation so that ingrowth of further high energy γ-emitters from the decaying U^{232} is minimised. The problem of the U^{232} decay chain hazard has been assessed by Arnold [8].

3.1. — Specification of Activity of Fuel.

The activity of the reference equilibrium fuel after four years irradiation and 200 days cooling has been calculated by F. P. O. Ashworth of Dragon Project. Table 2 shows the amounts of the principal β- and γ-emitters of interest in reprocessing in the reference plant and also summarises the γ-activities in curie MeV.

For remote refabrication a decontamination factor (DF) of 10^3-10^4 on gross γ-activity was specified for the solvent extraction plant, whilst for direct refabrication a DF of at least 10^6 is required.

Consideration is being given to the question of whether plutonium (arising from U^{234} in the 93 % enriched U^{235} make-up in refabrication) should be recycled or extracted. The issues involved are those of health physics, criticality control and the behaviour of Pu carbide during the heat treatment of the refabricated fuel kernels. In order to compare costs and to provide answers to the two first-named aspects pending experimental investigation of the last it was decided that Pu would be recycled in the minimum decontamination-remote refabrication route but that it would be removed for the maximum decontamination-direct fabrication.

Both decontamination routes, described below, were proposed by G. D. C. Short of UKAEA (Production Group, Windscale).

3.2. — The Intermediate Decontamination Route.

The flow diagram of the proposed "minimum" decontamination route is shown in Fig. 2.

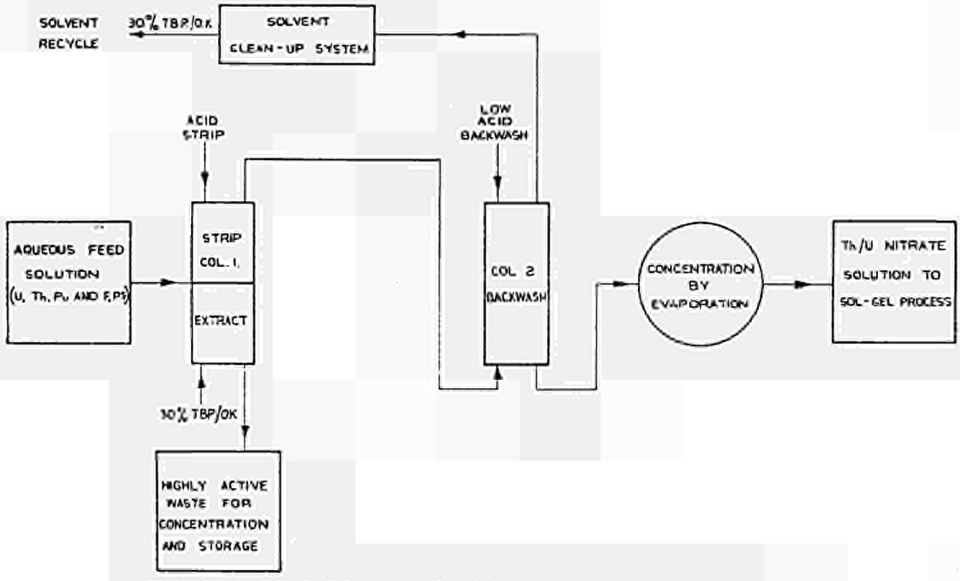

Fig. 2. — Block Diagram of Minimum Decontamination Process.

The separation process comprises the following principal stages :

1. A single solvent extraction column 1 in which the 0.1 M acid-deficient feed from the head-end process is contacted with a 30 % solution of TBP in odourless kerosene. A strip feed of 1 M HNO_3 is fed to the upper section of the column. The required decontamination factor is largely attained in this column, producing a raffinate which goes to the high activity waste treatment plant, described under 4.
2. The decontaminated U,Th product from column 1 passes to a back-wash column 2 where the U + Th are recovered as a slightly acid (0.08 M HNO_3) solution of nitrates.
3. The dilute U,Th product from the back-wash column is conditioned to a 0.12 M U/1.0 M Th solution containing 0.53 M free HNO_3 by steam stripping and evaporation. A volume reduction of about 86 % is required. The residual γ-activity due to fission products (2.5×10^4 mc MeV per day) is approximately equal to the γ-activity of the U^{232} decay chain remaining in the U, Th feed to the refabrication plant.
4. The aqueous raffinate from the fission product extraction column 1, containing one million curies per day of gross fission products, is concentrated to about one tenth of its initial volume so that high activity waste tankage requirements are reduced to about 67 litres per day's throughput of the reprocessing plant. The raffinate is steam-stripped to remove traces of TBP which interferes with the concentration process and increases the precipitation of solids in the high activity product. The 1.4 N liquor so produced is concentrated under vacuum and, in part, by evaporation by decay heat in the HA product tanks where it is stored at an acidity of 3-4 M HNO_3.
5. The contaminated TBP solvent raffinate from the back-wash column 2 is purified by a single wash with Na_2CO_3 solution at 60° C in a specially designed contactor. The solvent is then purified by steam flash distillation before being recycled to the decontamination column 1.

3.3. — The High Decontamination Route with Separation of Uranium from the Thorium.

For the direct refabrication of fuel a gross γ-decontamination factor of at least 10^6 is required. To achieve this level of activity the extraction system comprises the primary decontamination column 1 already described followed by the additional stages summarised below. Fig. 3 shows the flow diagram of this "maximum" decontamination process :

1. The organic phase from the fission product decontamination column 1 passes to a second column 2 where Pu is stripped back from the solvent by reduction to the (III) valency state with ferrous sulphamate. A further stream of 30 % TBP/OK is required at this column to strip U + Th from the aqueous Pu raffinate, which will be stored until sufficient quantity has been accumulated to warrant Pu recovery and utilisation.
2. After an acid back-wash in column 3 to return U/Th to the aqueous phase thorium is separated in column 4 by extraction with a 5 % TBP/OK solvent. The aqueous

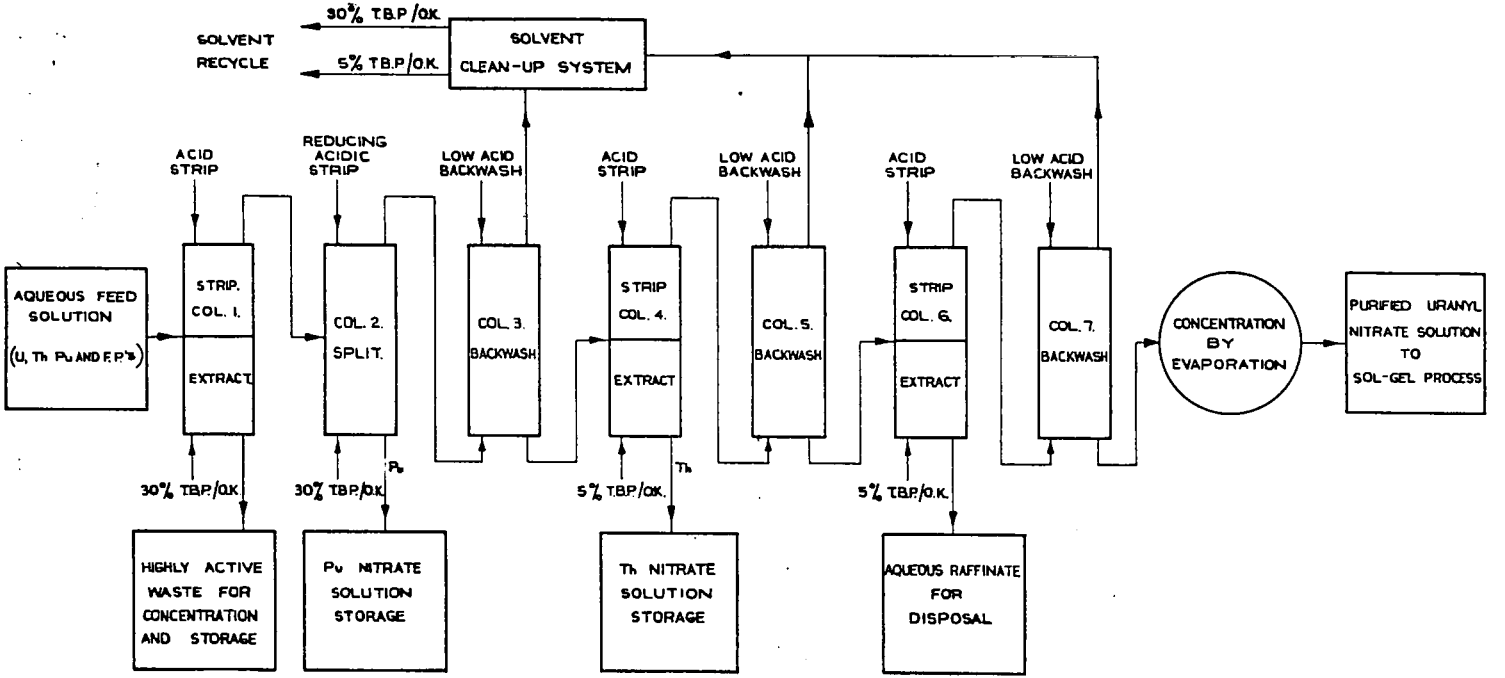

FIG. 3. — Block Diagram of Maximum Decontamination Process.

thorium raffinate is steamstripped and concentrated by evaporation to half its initial volume, producing 292 litres/day of approximately 1 M thorium solution for which storage is required for a decay period of about 15 years. Further decontamination of the uranium stream from fission products also occurs in this stage.

3. The uranium in the overhead product from the second cycle extraction column 4 is back-washed with 0.01 M HNO_3 in column 5 and evaporated to about one seventh of its volume. A uranium product storage tank is provided after this section.
4. A third and final extraction column 6 fed with 5 % TBP/OK reduces the residual fission product γ-activity of the uranium solution from column 5 to the desired level of less than 20 mc MeV/day.

   Uranium containing washing arising from decontamination of vessels and uranium solutions which may be held up during fuel fabrication are recycled to the product storage tank of column 5 so that in-grown activity of the U^{232} decay chain can be removed in this final decontamination cycle immediately before refabrication. The final solvent cycle thus combines two functions : the removal of fission products and thorium immediately prior to direct refabrication.
5. A final 0.01 M HNO_3 back-wash column 7 returns the clean uranium to the aqueous phase. A concentration factor of 13 is then achieved by evaporation yielding a 1 M solution of uranyl nitrate in < 2.1 M HNO_3 of the required low activity for refabrication. An overall γDF of 10^7 is attained.
6. Treatment of the high activity liquor and solvent purification are as for the minimum decontamination route. However, since two strengths of TBP/OK solvent are used here it is proposed that purification of each be carried out consecutively in a single flash distillation unit.

4. — The Refabrication of HTGR Fuel Uranyl Nitrate and Uranyl Nitrate/Thorium Nitrate Solutions

In the field of particulate Th/U fuel fabrication both the powder metallurgical and sol-gel methods have been operated in the Dragon Project. Much of the basic work on the powder route was carried out for Dragon by Huet and his collaborators at CEN, Mol, and similarly, basic and development work on the sol-gel processes applicable to the preparation of Th/U fuels is at present being undertaken on behalf of the Project at KEMA, Arnhem and CNEN, Rome. In addition further evaluation and development studies of both methods have been undertaken by Dragon Project staff at AEE, Winfrith. The principal objective of this work, as dictated by physics, geometrical and chemical requirements, has been the production of spheroidal porous hyperstoichiometric $\sim$ (Th, U) $C_{2.4}$ kernels of ~ 0.5 mm diameter in which the Th/U^{235} atomic ratio is 10.

As the initial charge for the Dragon Reactor Experiment was fabricated from 93 % enriched U^{235} the Th/U total ratio was 9.3 : 1 and most of the work both on the powder and sol-gel routes has been carried out with this composition. However,

provisional estimates of the composition of recycled fuels suggest that these ratios will change to a value of Th/(U^{233} + U^{235}) of 12 and a value of Th/U total of 7, the difference being due to the build-up of non-fissile U^{234} and U^{236} in the fuel.

4.1. — THE DRAGON POWDER METALLURGICAL METHOD FOR THE PREPARATION OF (TH/U) FUEL.

After a number of possible methods for the manufacture of (Th, U) C_ε kernels had been examined it was decided to employ one that involves the carbothermic reduction of urania and thoria.

The process described briefly below was developed after intensive experimentation and is now regularly used to produce porous sintered (Th/U) carbide fuel kernels.

The raw materials are :

(i) Urania : 93 % enriched in U^{235}, prepared by decomposition of ammonium diuranate treatment in hydrogen at 800° C and passed through a 300 micron sieve to remove coarse agglomerates.

(ii) Thoria : Powder passing a 37 micron (μm) sieve.

(iii) Carbon : United MT carbon black treated at 2,100° C in an argon stream to remove volatiles (BET = 6 m^2/g, particle diameter $\sim$ 0.5 μm).

(iv) Liquid paraffin BP, Δ = 0.865-0.890 g/cm^3.

The carbon, urania and thoria powders are dry-mixed in appropriate proportions to produce carbide kernels of the required chemical composition.

To this dry mixture is added paraffin as binder at a concentration of 6-8 cm^3/100 g of mixture which is then dispersed evenly throughout the mixed powders.

The damp mix is consolidated either by pressing or extrusion and the consolidated material granulated to produce crumbs of a size slightly smaller than the desired size of green kernel. These crumbs are then spheroidised by abrasion and "snowballing" in a Fritsch "Pulverisette" planetary mill. Green spheroids of the required size range are selected by sieving, the oversize being recycled through the granulation stage and the undersize being returned to the planetary mill.

Carbide kernels are formed by heating the green spheroids in vacuo at temperatures of up to 2,300° C. The porosity is controlled by varying the heat treatment, the higher temperatures being employed to produce denser kernels. After heat treatment, the sintered kernels are sized in an inert atmosphere and the appropriate size range sent for coating (Fig. 6). Outsize material and fines are oxidised and returned to the initial mixing stage.

This powder method is extremely flexible in that it requires only slight modification of technique to change the Th/U ratio or the carbon content of the final kernel. Both ThC_2 and UC_2 and many intermediate compositions have been made with this process. Furthermore, up to 30 % of carbon in excess of that required for stoichiometry can be incorporated into the kernels, which may be produced in sizes varying from 1.5-0.3 mm with porosities of between 10 % and 45 %. From the production aspect the principal time consumer is the granulation step and suitable equipment has still to be developed to carry this out on a scale that would be required for the reprocessing plant. By contrast the capacity of a Pulverisette type spheroi-

dising mill is very large. Rounded green kernels can be produced in a tight size range, e.g., — 572 μm + 500 μm at the rate of 5 kg/h with 10 kg/h of outsize material, which is recycled (Fig. 10). By widening the acceptable size range, considerably greater outputs may be obtained.

The loss of weight on heat treatment to carbide has been observed to be between 22 % and 23 %, a significant proportion being, of course, due to the paraffin binder.

No loss of fissile or fertile material during heat treatment has been detected by subsequent chemical analysis and 98-99 % overall yields of acceptable sintered kernels (— 500 + 300 μm) been obtained.

The properties of typical sintered kernels produced by this process are illustrated by the data tabulated below which were obtained from the first five batches of material to be prepared for the initial charge of the Dragon Reactor Experiment.

Properties of Sintered Fuel Kernels (—500μ +300μm)

Batch No.	Bulk Density g/cm³	Hg Density g	Gas Density g	Open Porosity %	Closed Porosity %	Th/U^{235}	Composition
1	3.22	5.89	8.65	31.9	3.0	11.04	$(Th, U)C_{2.49}$
2	3.3	6.63	8.59	22.8	3.8	10.5	$(Th, U)C_{2.51}$
3	3.47	6.22	8.4	25.9	4.1	10.1	$(Th, U)C_{2.62}$
4	3.65	6.86	8.6	20.2	4.6	10.2	$(Th, U)C_{2.44}$
5	3.16	5.83	8.7	33.0	3.3	10.0	$(Th, U)C_{2.42}$

At present fuel output in the Dragon Project fuel laboratory is limited by criticality control and furnace capacity. Both aspects need evaluation before scaling up to the throughput required by a reprocessing plant is feasible. Relatively little work has yet been carried out on the preparation of (ThO_2/UO_2) oxide kernels by the powder route, but preliminary results indicate that only minor modifications will have to be made to the process.

4.2. — Studies in Dragon Project of the Preparation of $(Th, U)C_2$ and $(Th, U)O_2$ Fuel Kernels by the Sol-Gel Process.

The manufacture of fuel kernels by the sol-gel technique has been undertaken in a number of laboratories and different approaches employed to meet their different requirements. Thus, at ORNL [9] there was interest in thoria containing a rather low percentage of urania; at CNEN, Rome [10] initial interest was in the preparation of pure UO_2 and at KEMA, Arnhem, preparation of microspheres of UO_2/ThO_2 with a mole ratio of 1 : 6.67 was undertaken [11]. From the aspect of refabrication of recycled fuel there are, no doubt, advantages and disadvantages in each of the routes developed by these three laboratories and most of the work in the Dragon

laboratories has been directed to combining aspects of all known processes to achieve a flexible and simple route for the production of porous oxide and carbide fuel kernels from recycled fuel.

4.2.1. — *The Sol-Gel Process Used in the Dragon Fuel Development Laboratory for the Preparation of Porous Oxide and Carbide Fuel Kernels.*

The process which is given in some detail below has been obtained by combining the chemistry of the KEMA sol-gel route with certain aspects of the technology of the ORNL sol-gel process. By doing this it has been possible not only to obtain satisfactory fuel kernels for irradiation experiments but also to developed a possible sol-gel route for the refabrication of Th/U dicarbide fuel kernels from a highly decontaminated uranium solution.

Preparation of (Th/U) Sols.

The preparation of (Th/U) sols is carried out by peptising freshly prepared $Th(OH)_4$ with uranyl nitrate and free nitrate ion at 100° C. This method, originally investigated at KEMA, is extremely simple and effective and produces very stable sols. The properties of such sols are however greatly influenced by the nitrate and uranyl ion concentrations (Figs. 4 and 5). Thus at ratios of $\frac{NO_3}{U + Th} \geqslant 0.4$, concentration of a 10 : 1 (Th/U) sol causes precipitation, whilst at ratios of $\frac{NO_3}{U + Th} \leqslant 0.3$ concentration merely increases the viscosity and the material becomes difficult to handle at high molarities (6 M-8 M). Similarly, when the $\frac{NO_3}{Th + U}$ ratio is kept constant at 0.4 there is no precipitation on concentration for Th/U ratios $\geqslant 20 : 1$ but precipitation on concentration occurs at Th/U ratios of $\leqslant 10 : 1$. However, for all Th/U ratios of interest to Dragon in the reprocessing of power reactor fuel, i.e., up to a uranium concentration corresponding to the Th/U ratio of 7 : 1, very stable sols can be prepared which do not precipitate when concentrated provided that the $\frac{NO_3}{Th + U}$ ratio is kept in the range 0.25 to 0.3.

Incorporation of Carbon into a (Th/U) Nitrate Stabilised Sol.

The most convenient method of incorporating carbon black into a (Th/U) sol is by violent agitation at room temperature, but before this is carried out present practice is to adjust the sol molarity (~ 4 M) by evaporation to the theoretical density of the carbon to avoid settling. Moreover, only certain carbons are suitable for adding to (Th/U) sols, critical parameters being the pH and the ultimate particle size of the material. However, sufficient carbon of the right quality can be incorporated into sols of this concentration to produce on heat treatment carbide kernels containing substantial quantities of free carbon [$(Th, U)C_{2.6}$].

When the sol is to be subsequently converted to porous oxide fuel kernels, carbon is also suspended in it to act as a temporary filler, but the concentration used is usually substantially lower than that used for preparing carbide fuel.

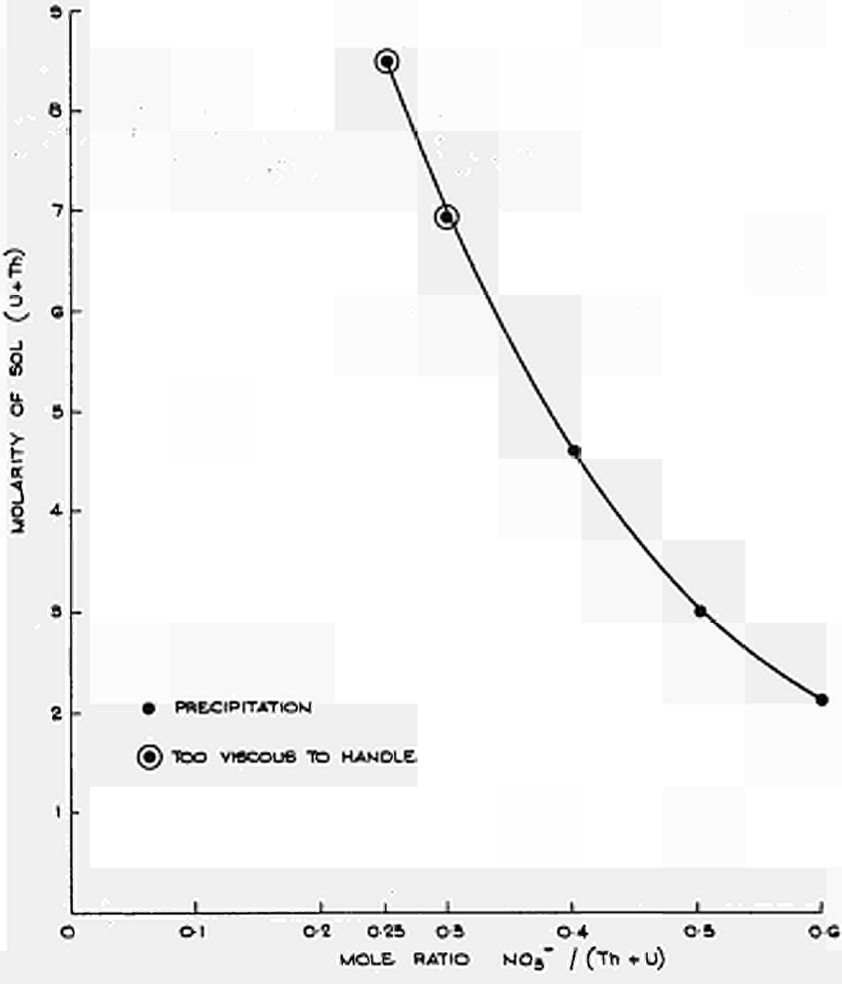

FIG. 4. — Maximum Molarity of Sol as function of $\frac{NO_3^-}{Th + U}$ Molar Ratio for $\frac{Th}{U} = 10$ Atomic ratio.

The Preparation of Gelled Microspheres.

To produce oxide and carbide fuel kernels of about 0.5 mm diameter from a suspension of carbon in a 4 M (Th/U) sol, the liquid is converted into droplets of $\sim$ 1 mm diameter suspended in 2-ethyl hexanol by the method developed at ORNL [9]. However, the gelling mechanism employed depends on the ratios of $\frac{NO_3}{Th + U}$ and

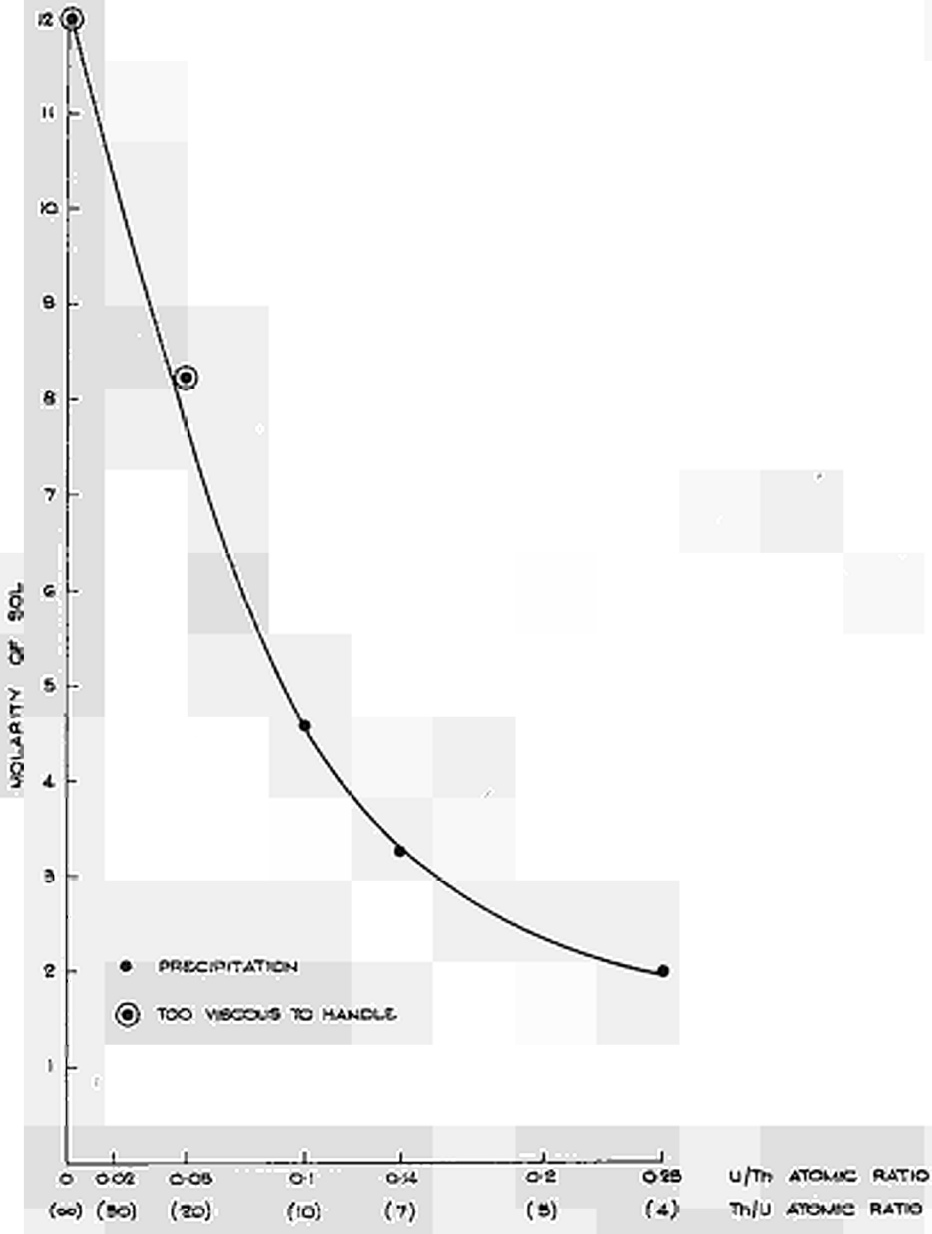

FIG. 5. — Maximum Molarity of Sol at 20° C as function of $\frac{Th}{U}$ Atomic Ratio

$$\frac{NO_3^-}{Th + U} = \text{Constant} = 0.4.$$

U/Th in the sol. If these are large enough to initiate precipitation when the sol is concentrated, then gelation is carried out internally by raising the pH of the sol with decomposing hexamethylene tetramine (1). The amine is added to the sol just before it is converted to droplets and its decomposition is effected by heating the hexanol (which must be nearly saturated with water to avoid precipitation) to 40-45° C.

On the other hand, if dehydration of the sol does not cause precipitation, the droplets can be gelled by dehydration in dry 2-ethyl hexanol. With both methods of gelation, it is usual to allow the spheres to remain in the gelling medium for 5-10 min before they are removed for further treatment. Furthermore both gelation methods require the presence in the 2-ethyl hexanol of a small concentration (0.2-0.3 %) of tensioactive agent (Span 80) to prevent coalescence of the droplets.

Treatment of Internally Gelled Microspheres.

The decomposition of hexamethylene tetramine in nitrate ion stabilised Th/U sols produces ammonium nitrate which, if present in too high a concentration, ruptures the spheres when they are heat treated. This is prevented by washing the gelled kernels, first in carbon tetrachloride to remove the 2-ethyl hexanol and then in 2 M ammonia solution which leaches out the ammonium nitrate. A final rinse with acetone removes the surface water from the kernels an and enables them to be converted rapidly to a free flowing condition.

Heat Treatment of Sol-Gel Kernels.

After the kernels have received the following air drying treatment :

80° C for 2 h
120° C for 2 h
180° C for 2 h

carbide kernels are formed by heat treating them in vacuo, a typical firing cycle being :

R. T.-900° C	2 h
900-1450° C	1 h
1450-1700° C	0.5 h
1700-2000° C	0.5 h
2000° C	1 h
2000° C-R. T.	1 h
Total treatment time . . .	6 h

This treatment has been used to produce satifactory crack-free carbide kernels in the size range 0.25-0.8 mm diameter with porosities in the range 20-30 % (see Fig. 7). Lower or higher porosities can, of course, be obtained by increasing or decreasing the final soaking temperature.

(1) This method was initially proposed for Th/U/C sols and investigated on behalf of the Dragon Project by KEMA.

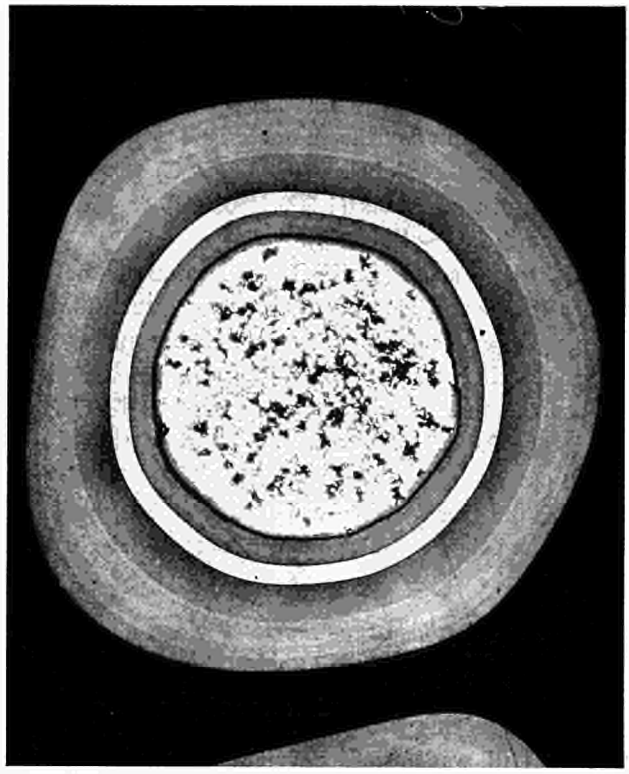

FIG. 6. — PyC/SiC/PyC Coated Sintered Carbide Kernel Prepared by Powder Metallurgical Route
Porosity = 36 % ×125

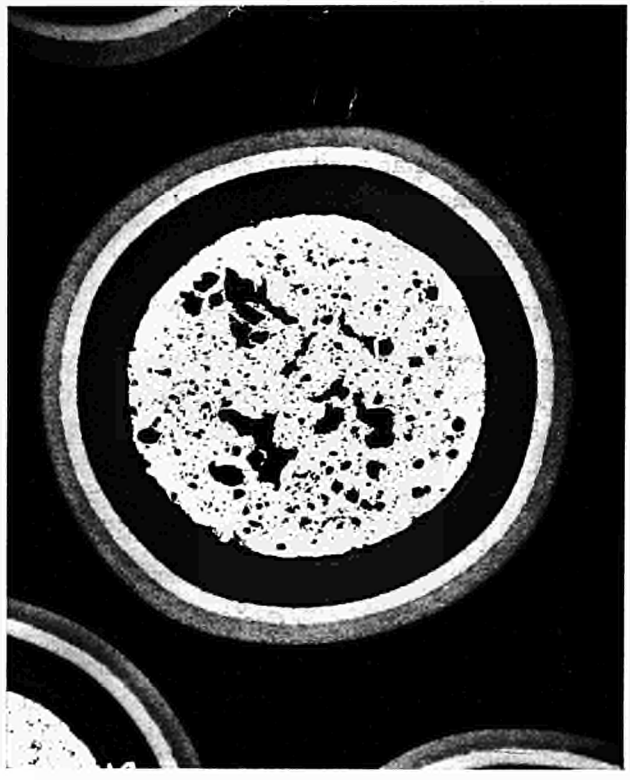

FIG. 7. — PyC/SiC/PyC Coated Sintered Carbide Kernel Prepared by Sol-Gel Route.
Porosity = 27 % ×125

To produce porous oxide kernels the heat treatment is entirely different and is designed to counteract the characteristic property of sol-gel oxide which is to densify at temperatures well below ($\sim$ 1,150° C) those usually employed for either heat treating thoria artefacts (1,600-1,800° C) or for coating with pyrolytic carbon or silicon carbide ($\sim$ 1,500° C).

Thus, to form the $(Th/U)O_2$ solid solution, the $ThO_2/UO_2/C$ dried green kernels are heated to a considerably higher temperature than that to be used in the subsequent coating process under a pressure of carbon monoxide which prevents the entrapped carbon reacting with the oxide. The kernels are then heated in an oxidising atmosphere to burn out the carbon. Very satisfactory $(Th/U)O_2$ solid solution porous kernels (size range 0.4-0.7 mm diameter) with extremely well distributed porosity (Fig. 8 and 9) can be made in this way. Porosities have so far been obtained in the range 20-60 % and this can be varied either by using different concentrations of carbon filler in the sol or by subsequent heat treatment of the decarbonised kernels.

4.2.2. — *An Outline of the Sol-Gel Process under Development for Dragon Project at CNEN, Casaccia (Rome).*

Although this process, the basic principles of which have previously been reported [10], was initially developed for preparing urania fuel kernels, it was considered that if it could be adapted to Dragon requirements it would be the most appropriate sol-gel route for converting a combined solution of thorium and uranyl nitrates from a reprocessing plant to carbide fuel kernels.

Preparation of Th/U/C Sols.

The thorium/uranyl nitrate solution ($\sim$ 1 M) is contacted in a mixer-settler with a long chain amine (Primene JMT) diluted (1 : 1) with white spirit, and as the $\frac{NO_3}{Th + U}$ ratio is slowly reduced a (Th/U) sol is formed. The extraction of nitrate ion is discontinued when the $\frac{NO_3}{Th + U}$ ratio has been lowered to a value of about 0.7 since further extraction causes the precipitation of UO_3. The sol is then concentrated by evaporation to about 4 M. To obtain superior gelation characteristics the sol may be subsequently treated with hydrogen in the presence of a catalyst to convert the uranium component from the hexavalent to the tetravalent state. However, the necessity for carrying out this treatment is still being appraised.

The sol is then mechanically blended with the appropriate quantity of carbon black that will produce fuel kernels of the required composition.

Conversion of Th/U/C Sol to Carbide Fuel Kernels.

Droplets of Th/UC sols prepared by amine denitration can be gelled within 5-15 min by dehydration, by further denitration with Primene JMT if the uranium component is in the tetravalent state, or internally, by the addition of hexamethylene tetramine. This operation is carried out in an ORNL [9] type fluidising tower using

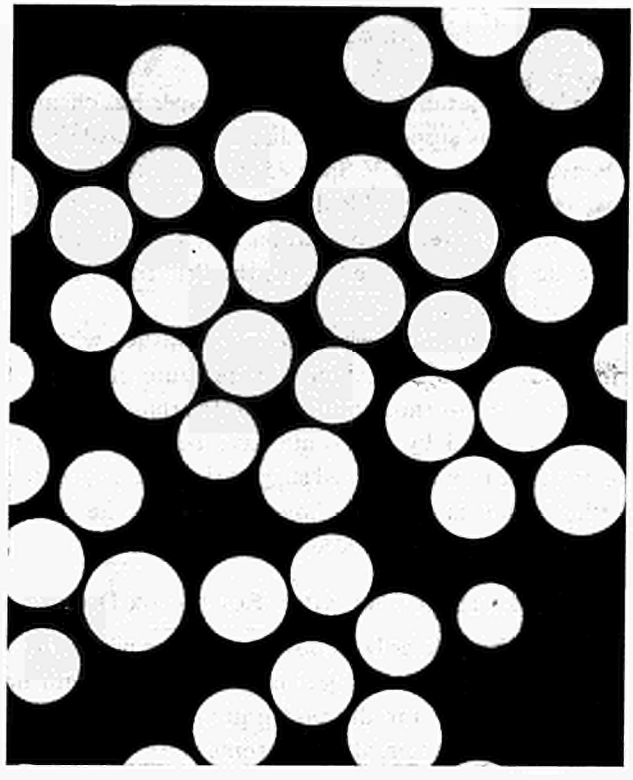

FIG. 8. — $(Th/U)O_2$ Sintered Porous Kernels Prepared by Sol-Gel Route.
(Th/U) = 10, Porosity = 31 %
Direct Illumination ×30

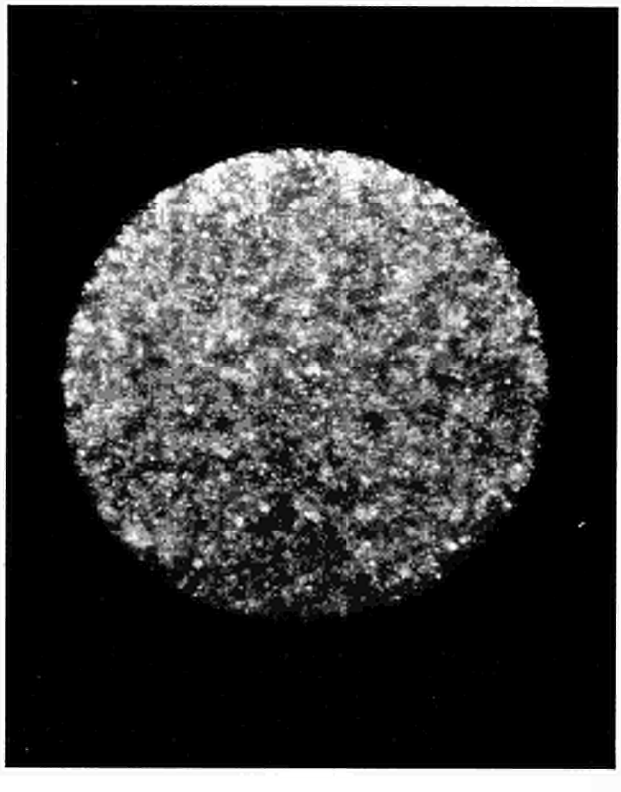

FIG. 9. — $(Th/U)O_2$ Sintered Porous Kernels Prepared by Sol-Gel Route.
(Th/U) = 10, Porosity = 31 %
Polarised Light ×125

either 2-ethyl hexanol or the much cheaper but chemically similar industrial solvent Alphanol 79 as suspension media. To avoid coalescence of the microspheres a tensioactive agent ($\sim$ 0.3 % Span 85) is added to the suspension medium when gelation is being produced by dehydration or by the addition of hexamethylene tetramine to the sol. However when gelation is caused by denitration, the presence of Span 85 in the alcohol is not necessary as the Primene JMT denitrating agent also prevents drop coalescence.

After gelling, the fuel particles are washed with carbon tetrachloride and slowly dried in air up to a maximum temperature of 425° C. Exploratory experiments on the conversion to thorium/uranium dicarbide at 1,800° C. in vacuo of dried sol-gel kernels produced by this route have been carried out by Van Geel [12] at CEN, Mol, and the results obtained suggest that a satisfactory product can be manufactured. Further study is required however, before the optimum heat treatment cycle is evolved for kernels produced in this route.

4.3. — Possible Refabrication Routes for Decontaminated Fuel.

As has been described above, experience within the field of particulate fuel manufacture in the Dragon Project has been gained with both the powder metallurgical and sol-gel routes for the preparation of, principally, porous carbide fuel kernels of $\sim$ 0.5 mm diameter, although some work has also been carried out on preparation of porous oxide fuel kernels of similar dimensions.

The choice of method for refabrication of recycled fuel will depend upon the technical and economic feasibility of adapting either of these routes to radiation conditions imposed by recycling decontaminated uranium and thorium solutions. Experience at ORNL [9] has already shown that it is possible to operate a sol-gel process in a shielded facility. Although similar experience with the powder metallurgical approach has not yet been obtained, it is felt that it should be possible to adopt this route also for fabricating fuel kernels behind shielding.

Thus, in the light of experience gained in the Dragon Fuel Development Laboratory and the experience contributed by the Project's collaborators at KEMA, Arnhem and CNEN, Rome, the fabrication routes shown in Fig. 10 are being considered for the production of recycled (Th/U) fuel kernels.

For the preparation of fuel kernels in the lightly shielded facility by the powder route, the uranyl nitrate solution must first be converted to urania powder and, since this is standard procedure in the fabrication of urania, this step should not be a major problem. The rest of the proposed route is identical to that at present in use in the Dragon Project Laboratories and requires the development of appropriate remotely controlled machinery to increase output to the level dictated by the throughput of the reprocessing plant.

The sol-gel route proposed for a highly decontaminated uranium stream is also essentially that at present employed in the Dragon Laboratories. It would, moreover, be feasible and, indeed, preferable to prepare the fresh thorium hydroxide outside the shielded facility and so avoid having to design it for the carrying out

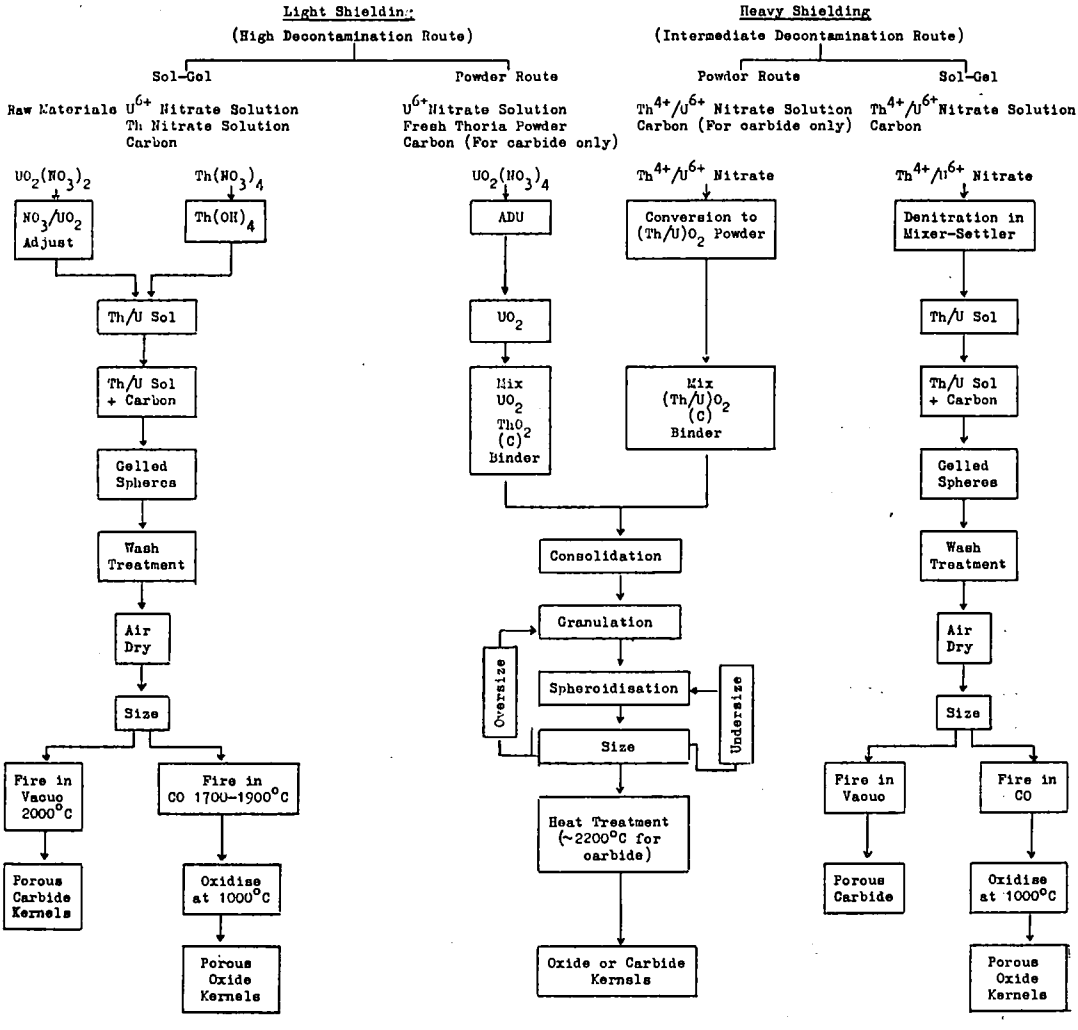

FIG. 10. — Refabrication Routes.

of quite a number of operations (dissolution, precipitation, filtration, ammonia wash and water leach) which produce considerable volumes of liquid waste. There may be, however, significant disadvantages with this approach in that the recycling of reject gelled, dried and fired materials may require different treatments. These aspects of the process are still being studied.

The use of the powder route in the heavily shielded facility will require the preparation within that area of large quantities of either $(Th/U)O_2$ solid solution or ThO_2/UO_{2+x} powder and as yet no work in the Dragon Project has been done on their preparation from a combined thorium/uranyl nitrate solution. This is not considered, however, to be a particularly difficult problem as many methods for

producing such materials have already been investigated [13]. Otherwise the fuel preparation route should be identical to that proposed for the lightly shielded facility.

For the preparation of fuel kernels in the heavily shielded facility from a combined uranyl nitrate/thorium nitrate solution by the sol-gel route, the method being developed at CNEN, Rome by Cogliati, *et al.* is considered to be the most appropriate and, indeed, it may also be considered for employment in the lightly shielded facility. Its great advantage is that the two components (Th and U) do not have to be separated which facilitates the recycling of reject material in that simple reconversion to (Th/U) nitrate solution is adequate.

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BURN-LEACH PROCESSES FOR GRAPHITE-BASE REACTOR FUELS CONTAINING CARBON-COATED CARBIDE OR OXIDE PARTICLES (1)

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ABSTRACT

Processing of graphite-base high-temperature gas-cooled reactor fuels containing carbon-coated carbide or oxide particles is discussed. The most promising process consists of burning the fuel in a fluidized-bed of inert alumina followed by leaching of the product bed with Thorex reagent to recover the uranium and thorium. Decontamination and final recovery of the uranium and thorium would be achieved by conventional solvent extraction methods. A preliminary engineering evaluation and potential costs are included.

1. — INTRODUCTION

High-temperature gas-cooled reactors (HTGR) that contain coated fuel particles dispersed in a graphite matrix are being developed. Eventually processes will be required for the recovery of uranium and thorium from these fuels. It is the purpose of this paper to summarize preliminary processing studies that relate to these fuels. Although materials such as pyrolytic carbon, SiC, ZrC, Al_2O_3, and BeO have been considered either as coatings or diluents for carbide and oxide fuel particles, this paper will be limited primarily to a discussion of processing methods for fuel that contains carbon-coated thorium-uranium carbide or oxide fuel particles. A promising processing method consists of burning the fuel in a fluidized bed of inert alumina and then leaching with fluoride-catalyzed nitric acid (Thorex reagent) to recover the uranium and thorium. Decontamination and final recovery of the uranium and thorium would be achieved by conventional solvent extraction methods. Laboratory- and engineering-scale studies of this process have been made with unirradiated prototype fuel specimens, and a few hot-cell experiments were run with irradiated material. The results of this work are summarized here; in addition, a preliminary engineering evaluation of the burn-leach process and costs as related to a conceptual fuel are included. An alternative method, burning followed by fluorination to recover the uranium as UF_6 is discussed briefly.

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2. — Burn-Leach Flowsheet

The burn-leach process for graphite-base fuels is shown in Fig. 1. Initially, the fuel is chopped or crushed and fed to a fluidized-bed burner. The best heat-transfer and fluidizing medium is probably a refractory grade of granular alumina. Design of the burner may be dependent on the type of fuel being burned. Burning at 700 to 750° C of fuels containing carbon-coated Th-U dicarbide particles converts the carbides to finely powdered oxides, dispersed homogeneously throughout the bed. Consequently, to recover the uranium and thorium, the entire bed must be leached. However, oxide fuel particles that have a high ThO_2 content might not be affected during combustion in a fluidized bed and probably can be separated from the alumina if desired before the leaching operation.

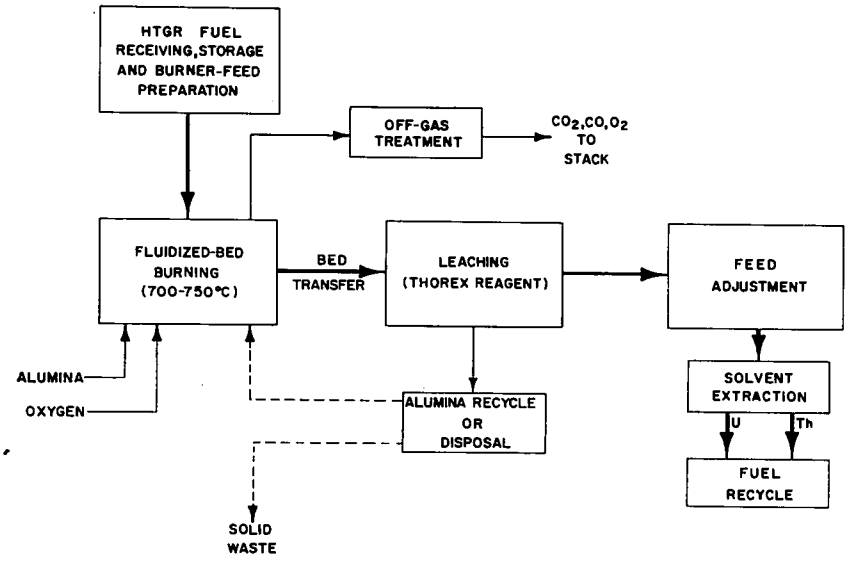

FIG. 1. — Potential Burn-Leach process for graphite-base reactor fuel.

Burning is started by injecting preheated oxygen into the fluidized bed and simultaneously heating the bed by external heaters. When the fuel starts to burn, the heaters are turned off, and the heat of reaction is removed by air-cooling the bed. For efficiency, continuous operation, with feeding of fuel, fresh alumina, and oxygen to the burner, and withdrawal of ash, all at the proper rates, is preferred. Toward the end of the reaction, when the amount of carbon in the bed has been reduced to a low level, the heaters must be restarted to complete the reaction. Under normal operation, nearly quantitative consumption of the oxygen is achieved, resulting in an off-gas composed mainly of CO_2. Carbon monoxide, which can form potentially explosive mixtures with oxygen, generally amounts to less than 5 % of the off-gas.

Particles in the off-gas are removed mostly by filters, and a gas-cleanup system prevents the release of all radioactivity except the noble gases.

After burning, the product bed is transferred to a leacher where the uranium and thorium are dissolved in fluoride-catalyzed nitric acid (Thorex reagent). The design of the leacher depends on the type of fuel being processed, since the product bed from carbide-bearing fuel will contain about 70 % Al_2O_3, whereas the product from oxide-containing fuel can consist mostly of ThO_2-UO_2 microspheres. In either case, leachates containing 0.5 to 1 *M* Th can be obtained. Less than 2 % of the alumina is dissolved; thus the remainder may be recycled or discharged to waste. Uranium and thorium recoveries should be greater than 99.5 %.

After the concentration of the leachate is adjusted, the uranium and thorium are separated from fission products and recovered by a conventional tributyl phosphate solvent-extraction process. The product from solvent extraction can probably be used as feed for the sol-gel process for refabrication of oxide or carbide fuel particles.

3. — Fluidized-bed Combustion of Graphite-Base fuel

3.1. — Fuels Containing Carbon-Coated Carbide Particles.

In the fluidized-bed burning of this type of fuel, the graphite is converted to carbon oxides, and the carbides to their respective oxides. The principal reactions are :

$$C + O_2 \rightarrow CO_2 \qquad \Delta H^o = -94.1 \text{ kcal}$$
$$2C + O_2 \rightarrow 2CO \qquad \Delta H^o = -52.8 \text{ kcal}$$
$$3UC_2 + 10O_2 \rightarrow U_3O_8 + 6CO_2 \qquad \Delta H^o = -1333 \text{ kcal}$$
$$ThC_2 + 3O_2 \rightarrow ThO_2 + 2CO_2 \qquad \Delta H^o = -434.7 \text{ kcal}$$

Tests of the combustion of fuel that contained carbon-coated carbide fuel particles were conducted in 2-in.-diam and 4-in.-diam fluidized beds, [1, 2] made of nickel. The smaller bed was used to prepare material for laboratory-scale leaching studies. The 4-in.-diam fluidized bed (Fig. 2) was used at Oak Ridge National Laboratory for pilot plant studies and had the essential features of a plant-size burner. It was divided into two sections by a perforated fuel-support grid located 2 ft above the bottom. The fluidizing gas, oxygen, was introduced through a ball-check gas distributor in the conical bottom; the ball-check also prevented plugging of the oxygen inlet line when the burner was shut down. Ash was withdrawn from the conical bottom through a cooled line by a small, 3/4-in.-diam, variable-speed screw conveyor. The bed was heated by resistance heaters and cooled by air. The enlarged disengaging space at the top was air-cooled and provided with internal fins to cool the combustion gases before filtration. Off-gas filtration was provided by two sintered stainless steel filters with an automatic blowback system. Each filter was 2-3/4 in. in diam, 18 in. tall, and had a 20-μ mean pore size. Crushed fuel and alumina were introduced into the bed via the disengaging section. The level of the fluidized

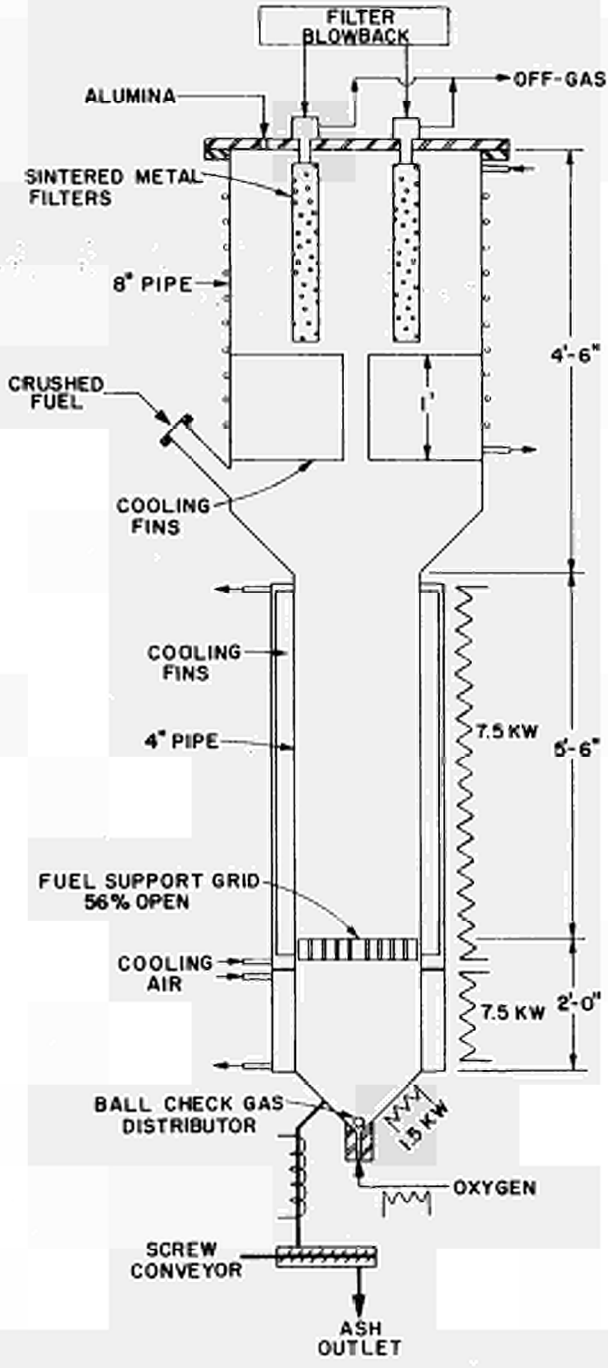

FIG. 2. — Four-inch-Diameter Fluidized-bed Burner for Pilot Plant Studies.

bed was determined with a thermocouple probe, and the behavior of the bed was observed through a sightglass in the center of the top flange during initial experiments with only alumina present. Other thermocouples and pressure taps were provided at various points for control purposes. The unit was made of nickel 201 so that halogens could also be used in it. Type 310 stainless steel would probably be preferred for a large-scale burner because of the better high-temperature mechanical properties of this material.

The fluidized bed of alumina was an efficient heat-transfer medium. Also, it prevented hot spots and resultant clinkering as it diluted and suspended the fuel oxides during combustion. As a result, the product was a free-flowing powder. Combustion was done at 700 to 750° C and was easily controlled. It should be pointed out that although the term "fluidized bed" is used, the entire contents of the bed were usually not fluidized by the gas stream. For instance, the larger pieces of graphite fuel formed a loosely packed bed, which rested on the fuel-support grid. Alumina fluidized in the gas passages through the bed and in the free space below and above the fuel. The best operation [1] was achieved when equal parts of 60- and 90-mesh Norton Abrasive Company Blue Label R.R. fused alumina was used. For example, when the initial alumina charge contained an appreciable amount of fines, as for example in a mixture of equal parts of 60-, 90-, and 120-mesh particles, the degree of fluidization decreased as additional fines were produced by the oxidation reactions, especially when the concentration of U-Th oxides approached 30 wt %. This eventually resulted in loss of fluidization, channeling, and hot spots. Satisfactory operation was restored by adding coarse alumina to regain the correct particle-size distribution for good fluidization in the presence of large amounts of fines derived from oxidation of the fuel. A one-week test showed that attrition of the fluidized alumina was negligible under normal operating conditions [2].

Chopped or crushed fuel was added to approximately 20 kg of alumina, and the bed was fluidized with preheated nitrogen while additional heat was supplied by the external heaters. Since the fuel ignites at 650° C, at this temperature the nitrogen was replaced by oxygen and the heaters were turned off. Bed centerline temperatures and wall temperatures were held at about 750 and 700° C, respectively, by air-cooling the finned exterior of the fluidized bed. The concentrations of CO_2 and CO in the off-gas were continuously monitored by gas-chromatographic and infrared analyzers. The CO_2 and CO contents of the off-gas were relatively constant when there was an excess of carbon in the burner; a decrease in the CO_2 and CO contents showed that the carbon inventory in the bed was being depleted. Accordingly, more chopped fuel was added as needed to maintain the desired oxidation rate. Alumina was added periodically when product was continuously withdrawn. The carbon inventory in the burner was normally 2 to 4 kg during a continuous run. The major part of the reaction occured in the bed of chopped fuel that rested on the support grid. Small particles of carbon tended to be elutriated to the region above the packed bed and reached an equilibrium concentration in the upper part

of the bed. Any small particles of carbon entrained in the alumina below the grid were rapidly burned in the hot oxygen, and it was possible to continuously withdraw an almost carbon-free product stream from the bottom of the bed. Toward the end of a combustion run, when the carbon concentration in the bed was low, it was necessary to supply heat to the burner to ensure combustion of the last traces of carbon. The superficial gas velocity in the bed was about 0.76 ft/sec at the bed midpoint pressure of 17.6 psia and average temperature of 725° C.

Continuous oxidation rates [1] varying from 1.1 to 1.4 kg of carbon per hour were obtained in pilot plant tests with a 4-in.-diam fluidized bed by varying the oxygen flow rate over the range of 1.3 to 1.6 scfm. Oxygen utilization decreased from 97 to 90 % as the flow rate was increased. Other variables affecting the rate of oxidation were : carbon inventory (or perhaps more correctly, carbon surface area) in the bed, operating temperature, and oxygen content of the feed gas. The average heat flux, based on the inside area of the reactor, was about 4,800 Btu hr^{-1} ft^{-2}, or about 2,100 Btu hr^{-1} ft^{-2} of outside surface on the finned outer wall. The heat transfer coefficient from bed to wall was estimated at 85 Btu hr^{-1} ft^{-2} °F.

The off-gases were cooled in the top of the reactor, passed through the filters at the top of the burner to remove entrained carbon dust and oxides, and sent to an off-gas system. Plugging of the filters was not a problem, and the filter blowback system was not used during routine operations. Micropore filtration [2, 3] of the off-gas for various periods showed that practically no particles escaped through the primary sintered-metal filters, thus demonstrating that the dust-coated filters were extremely efficient. Previous work [4] had shown that 99 % of the particles in fixed bed combustion off-gas were less than 0.3 μ in diameter. Oxygen utilization in the burner was high, as noted above, and a typical off-gas consisted of about 90 % CO_2, 5 % CO, and 5 % O_2. The CO concentration in the off-gas was below the flammability limit. Corrosion of the burner was negligible, [1] about 4 mils/year.

The carbon content of the discharged bed was generally less than 0.1 %. Beds containing up to about 30 % oxides ($ThO_2 + U_3O_8$) were produced [2], with no noticeable difference in operating efficiency.

3.2. — Fuels Containing Carbon-Coated Oxide Particles.

To date, no fuel containing carbon-coated ThO_2-UO_2 microspheres has been burned in a fluidized bed. A problem with this fuel arises from the uncertainties concerning the integrity of the microspheres after irradiation to projected burnups of 50,000 to 80,000 Mwd/metric ton of thorium plus uranium. If the microspheres retain their shape, it might be possible to separate them from most of the alumina after burning the carbon. This might result in a simpler leaching system. However, if the particles are broken during irradiation or combustion, they will be dispersed thoroughly in the alumina, requiring that an excess of alumina be fed to the leaching system. In either case, the same fluidized-bed burner probably could be used. A conceptual drawing of a large burner is shown in Fig. 3; it is a scaleup of the pilot

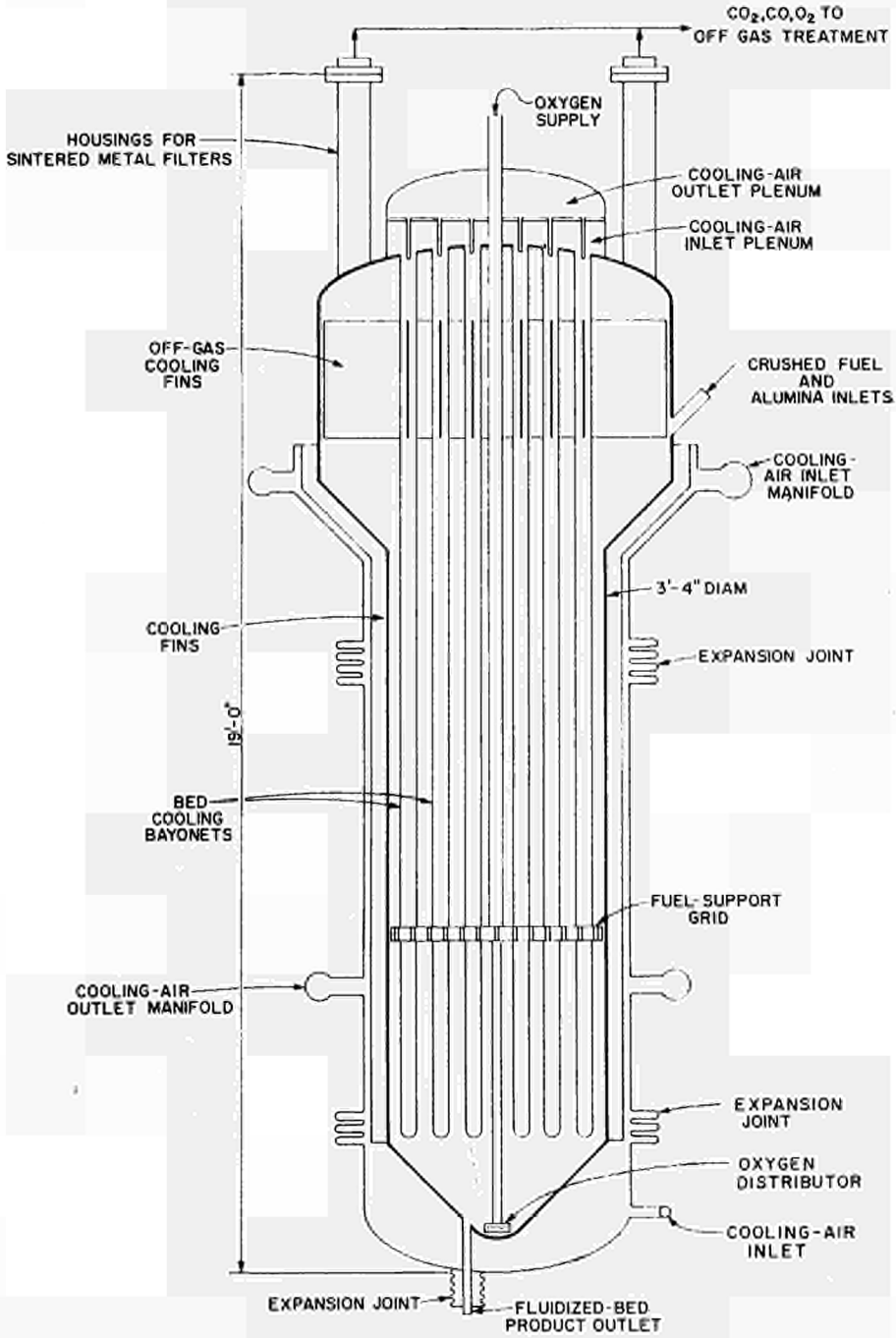

FIG. 3. — Conceptual Design for a Large, Fluidized-bed Burner for Graphite-base Fuels.

plant burner and would be operated similarly. Provisions for removing heat from more central portions of large fluidized beds must be made to avoid excessive centerline temperatures. This additional heat transfer surface is provided by air-cooled bayonets immersed in the bed.

For this study, it was assumed that the microspheres would not be broken and that a classifying operation after burning would permit leaching a product that is practically all microspheres with very little, if any, alumina. Eventually, the alumina will have to be discarded through the classifier and the leacher because of degradation of the alumina by attrition. However, as noted previously, the attrition seems to be very slow and the useful lifetime of the alumina is expected to be long.

3.3. — Fission Product Behavior During Combustion.

The behavior of the fission product was not studied during actual fluidized-bed combustion but was examined cursorily in laboratory-scale tube-furnace experiments. In one series of experiments [5], in which a large excess of oxygen was used to burn prototype Peach Bottom fuel irradiated to about 10,000 Mwd/metric ton (U + Th), up to 35 % of the cesium and 96 % of the ruthenium were volatilized from the high-temperature zone during 6-hr combustions at 800° C. Experiments [6] in the same equipment with a slightly irradiated fuel at 700° C showed that up to 1.1 % of the cesium and 65 % of the ruthenium were volatilized in 6 hr. In each case, practically all the fission products were trapped in the cool end of the reaction tube and nearly all remaining activity was removed by filtering the off-gas through a clean, sintered, 40-μ-porosity metal filter in a manner similar to the method proposed for a plant-scale fluidized-bed burner. The overall decontamination factor was greater than 10^4 in all experiments. In other studies [7] only a small amount of cesium and ruthenium was volatilized from the hot zone when the fuel was burned in a deficiency of oxygen at 800 °C. Waste calcination experiments [8] demonstrated the beneficial effect of a reducing gas (CO or NO) and elevated temperatures in decreasing ruthenium volatility by reducing higher oxides to the less volatile RuO_2. The preceding data suggest that volatilization of fission products, other than the noble gases, will not occur to a significant extent in a fluidized bed if the oxygen consumption is nearly quantitative and if the off-gases are cooled before filtration. Furthermore, if the ThO_2-UO_2 microspheres retain their integrity during irradiation and combustion, little release of activity to the fluidized bed is expected during combustion.

3.4. — Final Off-Gas Treatment.

Initial cleanup of the off-gas is achieved by cooling and then filtering it through sintered-metal filters in the burner. The gas passing through the filters (CO_2, CO, O_2, noble-gas fission products) should be nearly free of particles, as noted above. Iodine will not be present in significant amounts if the decay period is sufficiently long before it is processed. Supplemental treatment of the off-gas could be made in several ways, and several useful high-efficiency methods have been demonstrated in the waste calcination programs at Idaho [8] and Handford [9]. One method might

consist in passing the off-gases through a silica-gel trap, which would adsorb ruthenium (if present) and act as a deep-bed filter, then through absolute filters, and finally to a stack. However, this may not give sufficient decontamination. Scrubbing with acidic or caustic solutions could also be done, but this is not very efficient for trapping small particles [10]. Furthermore, scrubbing with caustic would result in the absorption of CO_2 and the generation of a large amount of aqueous waste. Significantly, scrubbing with caustic was not very effective in cleaning the off-gas from a waste-calcination process [9]. A more attractive method might be to mix steam with the off-gas, condense the vapor, and then filter through absolute filters. Waste-calcination work [9] indicated that a system combining sintered-metal filters, condensation of vapor, and finally absolute filtration can yield decontamination factor $\geq 10^8$ for the off-gas. When fluidized-bed burning tests are made with irradiated fuel, we will be able to define the off-gas cleanup problems more clearly and specify the cleanup system in more detail. However, at this point it does seem probable that a high-efficiency cleanup system which contributes a negligible amount to radioactive liquid and solid wastes can be devised for the fluidized-bed burning of graphite fuel.

4. — Leaching of Fluidized-Bed Products

4.1. — Products from Fuels that Contain Carbon-Coated Carbide Particles.

Since the product from the combustion of this type of fuel is a homogeneous dispersion of thorium and uranium oxides in alumina, the entire bed must be leached. In the first leaching tests [1, 2], a simple cylindrical vessel in which the bed was supported on a sieve plate was used. With this leacher, preheated leachant was circulated either upflow or downflow through the bed. A more efficient bench-scale batch leacher was devised [2] (Fig. 4); the leaching acid was recirculated upflow through the bed, fluidizing the bed. Excellent contact was obtained, and gas evolution and boiling in the bed did not interfere with the leaching. Then, the product solution was drained from the bed, and the bed was washed with water. In practice, the washed alumina would then be slurried with water and transferred to an alumina recovery system where it would be dried and then either discarded or returned to the burner.

Leaching resulted in uranium and thorium recoveries of greater than 99.5 %, while less than 2 % of the alumina was dissolved. The main reactions are :

$$U_3O_8 + 8HNO_3 \rightarrow 3UO_2(NO_3)_2 + 4H_2O + 2NO_2$$
$$ThO_2 + 4HNO_3 \rightarrow Th(NO_3)_4 + 2H_2O.$$

Very little heat is evolved by either reaction; this requires that the leachant be heated — to boiling if maximum efficiency is desired.

In cold bench-scale tests [3] with a bed consisting of 3 % U_3O_8, 17 % ThO_2, and 80 % Norton R. R. alumina, more than 99.9 % of both the uranium and thorium were recovered by leaching with 12 *M* HNO_3 — 0.04 *M* HF at 110° C. Laboratory-

scale leaching studies at Oak Ridge National Laboratory showed that excellent recoveries of uranium and thorium can be achieved with a variety of Thorex-type leachants. The bed material for these studies was produced by burning unirradiated Peach Bottom fuel compacts (carbon -coated Th-U dicarbide particles dispersed in a graphite matrix) in a fluidized bed of Norton R. R. alumina at 700 to 750° C. The product bed consisted of 6 % U_3O_8, 25 % ThO_2, and 69 % Al_2O_3. In 5-hr leaches, more than 99.5 % of the uranium and thorium were recovered when the HNO_3 concentration was 4 *M* or higher, and when the HF concentration was 0.02 to 0.05 *M* (Table 1). Aluminium nitrate, up to 0.1 *M* in the leachant, had no adverse effect on the recoveries. In no case was more than 2 % of the alumina dissolved. Uranium and thorium recoveries were inadequate with 13 *M* HNO_3 and with 2 *M* HNO_3 — 0.05 *M* HF (Table 1). The effect of lower leaching temperatures is being investigated.

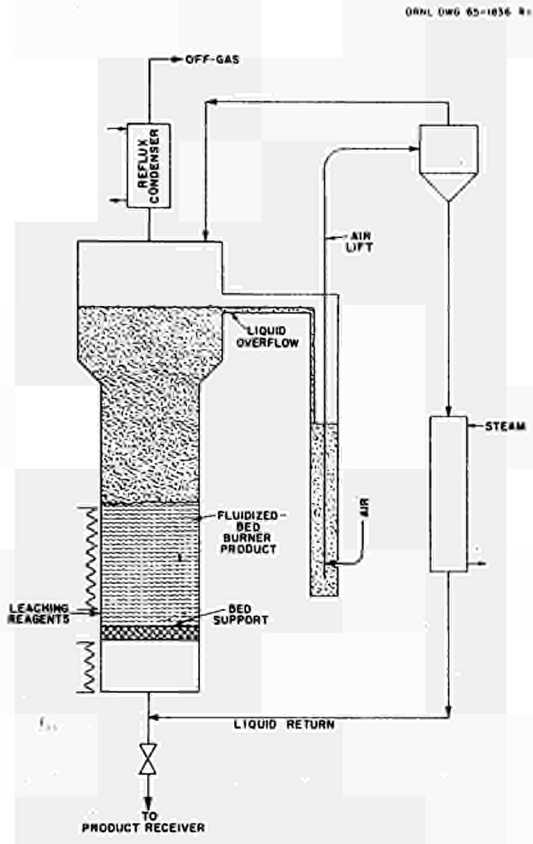

FIG. 4. — Bench-scale Batch Leacher.

4.2. — PRODUCTS FROM FUELS CONTAINING CARBON-COATED OXIDE PARTICLES.

As mentioned previously, there has been no experimentation with fuels that contain carbon-coated oxide particles. However, if this fuel burns as predicted,

TABLE I. — Results of Laboratory-Scale Leaching Experiments.
Leachant : Boiling HNO_3-HF-$Al(NO_3)_3$
Fluidized-bed product from combustion of Peach Bottom fuel :
6 % U_3O_8, 25 % ThO_2, 69 % Norton R. R. alumina
Leaching time : 5 to 7 hr
Final solutions were about 0.6 M in Th

Leachant Composition (*M*)			Amounts Leached (%)		
HNO_3	HF	$Al(NO_3)_3$	U	Th	AL
2	0.05	0	83.6	83.0	0.2
4	0.05	0	99.9	99.9	1.9
4	0.05	0.1	99.7	99.5	—
13	0.0	0	27.7	14.8	—
13	0.02	0	99.9	99.6	—
13	0.05	0	99.9	99.9	1.9
13	0.05	0.1	99.9	99.9	—

leaching of uranium and thorium may simply involve dissolution of ThO_2-UO_2 microspheres in the presence of a small amount of alumina. For criticality control, use of two geometrically safe slab leachers in series is envisaged for dissolving practically all the fuel particles before the solution and alumina slurry flow into large-diameter feed adjustment vessels. A schematic drawing of the leacher system is shown in Fig. 5. The leachers would be equipped with thermosiphon heating loops

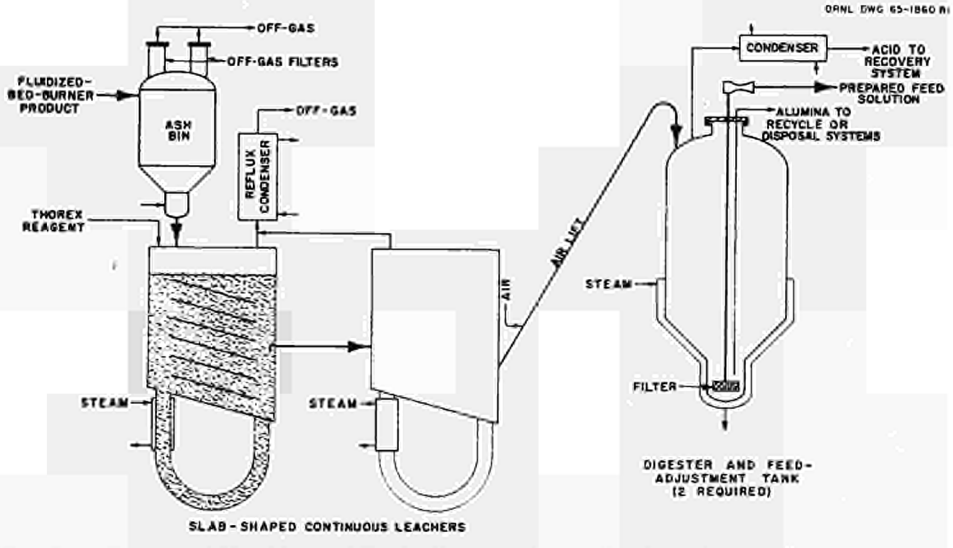

FIG. 5. — Conceptual Heaching and Feed-adjustment System for Graphite-base Fuels that contain ThO_2-UO_2 Microspheres.

and would operate continuously in series. Solids (mainly fuel particles still being dissolved and the insoluble alumina) that settle out on the sloping trays in each leacher would be partially recirculated within that leacher. Eventually the insoluble alumina would be transported through the system with the leachant to the feed adjustment vessels. Leachant would be pumped into the first slab-shaped tank and maintained at its boiling point throughout the leaching system. Holdup time in each leacher would be about 3 hr. Solids and solution from the first leacher would overflow continuously into the second one. Alumina would be transported through the system without being attacked appreciably by the dissolvent. Solution from the second leacher would be transferred semicontinuously by intermittent air lift to a feed adjustment system where any small fuel particles still remaining in the leacher overflow would be dissolved rapidly.

Unlike the oxide product from the combustion of Th-U dicarbide particles, unirradiated ThO_2-UO_2 microspheres probably cannot be dissolved readily in a dilute Thorex solution (Table 2). However, almost theoretically dense, 300- to 600-μ-diam ThO_2 microspheres were dissolved in 3 to 6 hr in boiling 13 *M* HNO_3-0.05 *M* HF, even in the presence of a large excess of alumina. In other studies [11, 13], irradiated ThO_2-UO_2 appeared to dissolve faster than unirradiated oxide. Thus, the 6-hr dissolution period estimated for fuels containing ThO_2-UO_2 microspheres should be adequate.

TABLE 2. — Dissolution of Sol-Gel ThO_2 Microspheres in Boiling Thorex Reagents
Reaction time = 6 hr

Diameter of Microspheres (μ)	Reagent Composition (*M*)		Amount Dissolved (%)
	HNO_3	HF	
250-300	2	0.05	20
420-600	2	0.05	33
250-300	4	0.05	57
420-600	4	0.05	73
250-300	13	0.05	100
420-600	13	0.05	100

5. — SOLVENT EXTRACTION

The solvent extraction system for recovering the uranium and thorium is based on the newer processes, [14] in which an acid-deficient feed is used for maximum fission product decontamination. Nitric acid is used as the salting agent instead of $Al(NO_3)_3$. This reduces the amount of $Al(NO_3)_3$ in the stored waste. If thorium recovery is desired, a process similar to the Acid Thorex Extraction process (Fig. 6)

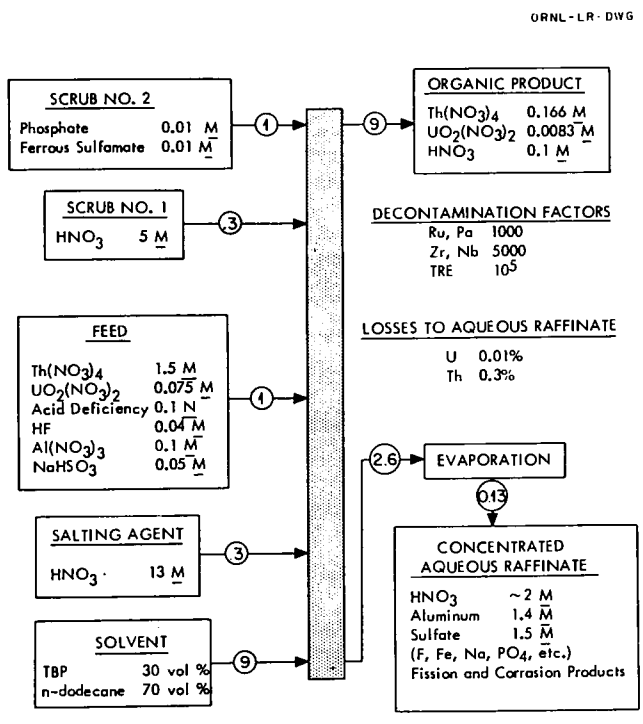

FIG. 6. — Acid Thorex Process for Co-extraction of Uranium and Thorium.

could be used. The organic product has a low acid content; consequently, the thorium and uranium can be co-stripped with a minimum aqueous volume and number of stages. Uranium and thorium may also be recovered separately in partitioning and stripping columns if desired. If thorium recovery is not desired, the Acid-Interim-23 process [14] could be used. The irradiated thorium will probably contain enough ^{228}Th to interfere with fuel manufacture in lightly shielded fuel-refabrication facilities. Consequently, until such time as heavily shielded facilities are available, it may be desirable to concentrate the thorium product and store it for about ten years to permit decay of ^{228}Th before recycle of the thorium.

6. — PRELIMINARY ECONOMIC EVALUATION

A preliminary plant design and capital- and operating-cost estimates were made for a head-end facility that would provide for irradiated fuel-element receipt and storage, crushing, burning, leaching, feed adjustment to acid-deficient conditions suitable for solvent extraction, adjusted feed-storage surge capacity, and tank storage of the recovered thorium for ^{228}Th decay. This head-end facility is assumed to be located at a conventional fuel-processing plant, such as the one built by Nuclear Fuel Services, Inc. (NFS). At this plant, the uranium and thorium would be recovered separately by solvent extraction, the thorium returned to the head-end facility for decay storage, and the fission product waste concentrated for disposal.

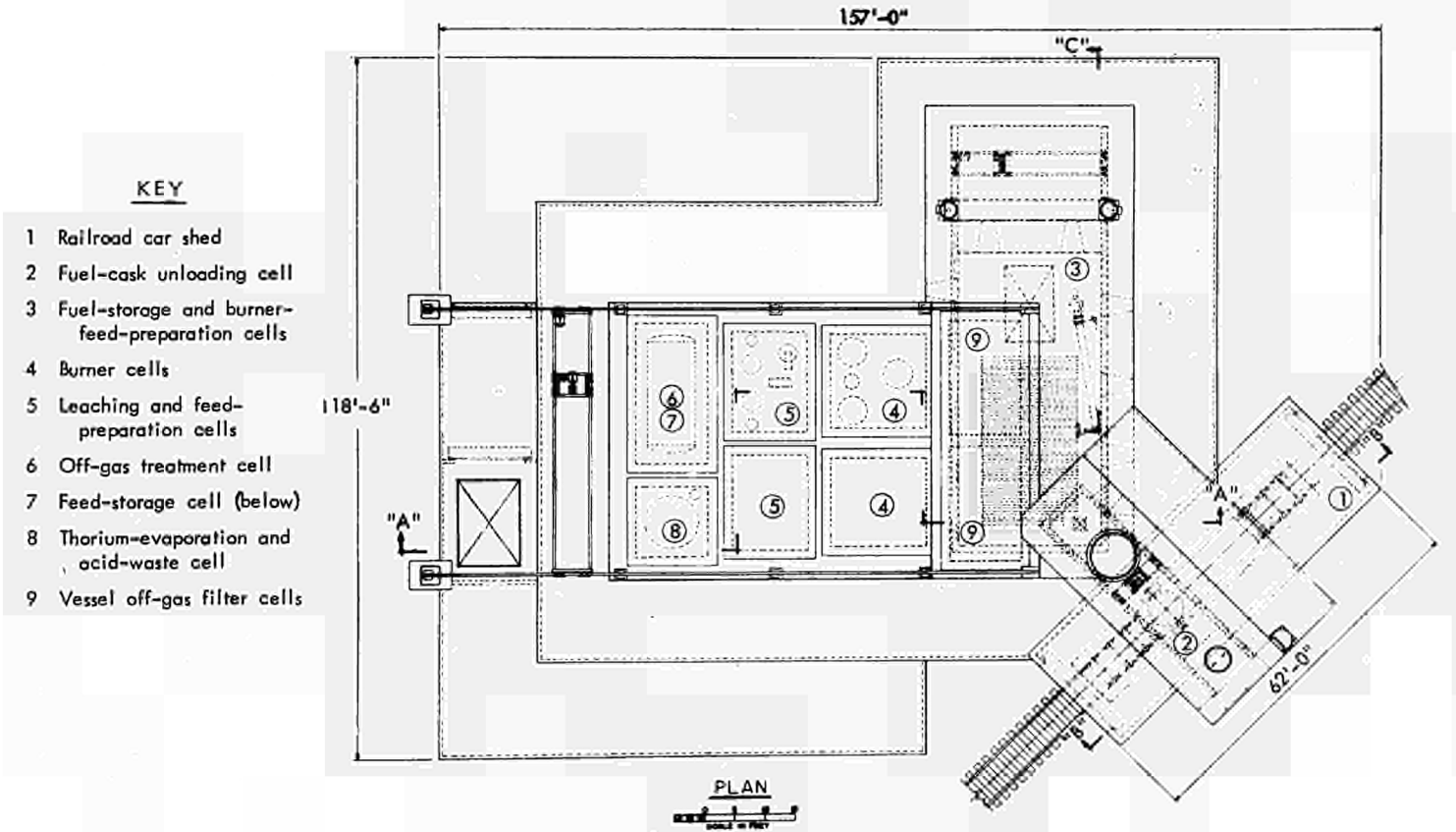

FIG. 7. — Fuel-reprocessing Head-end Building for HTGR Fuel.

KEY

1 Fuel-cask unloading cell
2 Fuel cans on storage rack
3 Fluidized-bed burner
4 Silica-gel adsorber for ruthenium
5 Waste-alumina bin
6 Ash bin
7 Continuous leacher
8 Digester and feed-adjustment tank
9 Thorium evaporator
10 Acid-waste-collection tank
11 Feed-storage tank
12 Vessel off-gas filter cell

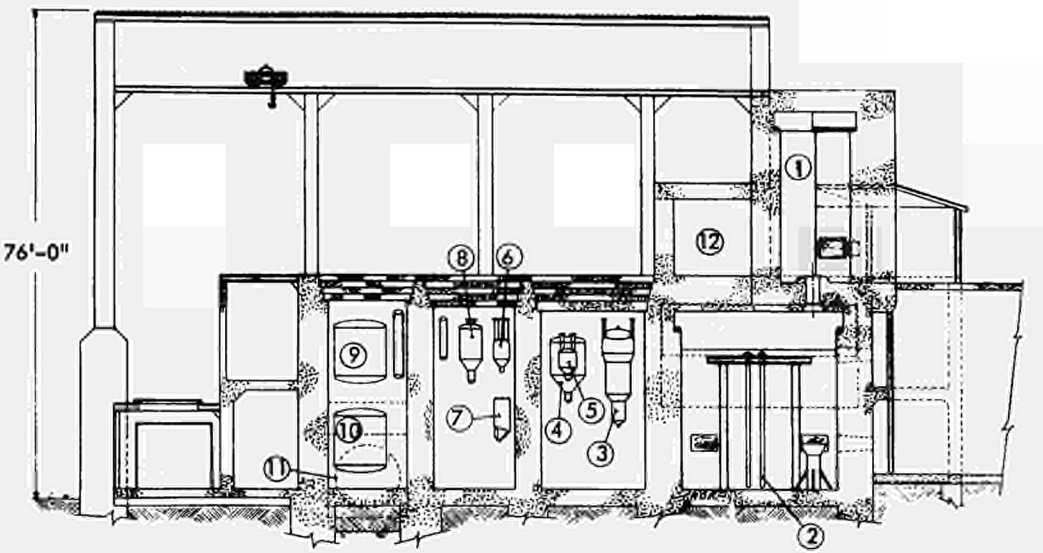

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FIG. 8. — Fuel-reprocessing Head-end Building for HTGR Fuel.

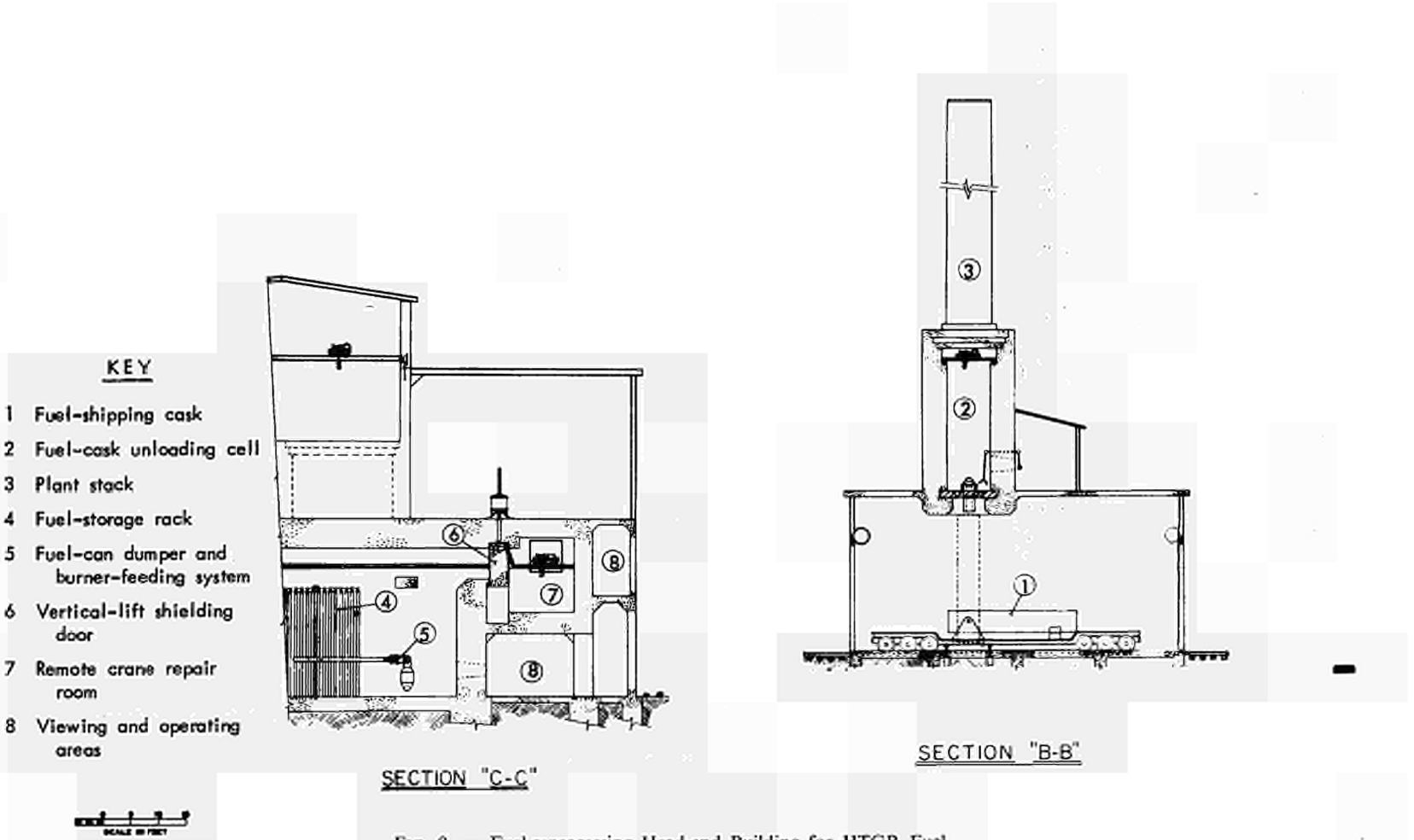

FIG. 9. — Fuel-reprocessing Head-end Building for HTGR Fuel.

The study was made for a fuel that contains sol-gel oxide microspheres coated with pyrolytic carbon, which may be the favored HTGR fuel of the future. Although the processing of carbide-particle fuels may be somewhat different, the resulting differences in estimated costs would probably be small and within the margin of error of the estimate. The fuel element is assumed to be a 4.5-in.-diam, 20-ft-long, graphite "log" from which the fueled particles are not easily separable; thus, provisions are made for crushing and burning the entire element. Each element contains 107 kg of carbon and about 10.9 kg of thorium plus uranium, before irradiation. After an assumed burnup of 50,000 to 80,000 Mwd/metric ton in about 6 years of irradiation, and 6 months or more postirradiation time for fission product heat reduction and protactinium decay, the element contains about 10 kg of thorium plus uranium, the balance having been converted to fission products.

The conceptual design of the head-end facility is shown Figs. 7, 8, and 9. The processing capacity is 40 elements per day for up to 225 days a year, with two parallel lines of crushing-burning-leaching-adjustment equipment (Figs. 3 and 5). At maximum throughput rate, such a facility could handle the fuel from reactors having up to 10,000 Mw (electrical) of installed capacity; however, as discussed below, the cost of the plant might have to be borne by a smaller industry than this. Fuel unloading and storage cells are designed for remote operation and limited remote maintenance, and the chemical process cells are designed for direct maintenance. Analytical, administrative, chemical supply, waste disposal, and plant utility services are assumed to be provided by the associated solvent extraction plant, with appropriate enlargements where necessary. A complete, independent, radioactive off-gas and cell-ventilation system, plus disposal stack, are included. Limited office and changeroom facilities are provided in the building. An 11,500-gal surge tank is included to hold up about 16 metric tons of thorium plus uranium as adjusted feed, permitting some scheduling leeway between the head-end and solvent-extraction facilities. An evaporator for the partially decontaminated thorium second-cycle raffinate is included, as is a 12,400-gal storage tank which holds about 33 metric tons of thorium as the hydrated nitrate. Additional thorium storage tanks would be built as needed, and these are assumed to be paid for by the value of the thorium when it is recycled after the ^{228}Th decays.

The estimated capital cost of this facility is $9,040,000 (Table 3). This includes $1,260,000 for the second processing line, an expenditure that could be postponed until required if the plant were started on a fraction of its design load. The "standby" operating cost, that is, the minimum labor, utility, and overhead cost of maintaining the facility when fuel is not being processed, is estimated to be $115,000 a year. Additional labor and overhead costs when fuel is being processed are estimated at $350 a day, The cost of oxygen and alumina is estimated at $846 a day for one burner line, or $1,020 for two burner lines. The lower unit cost for two lines is the result of lower oxygen costs at the high usage rates. The costs of nitric acid, other chemicals, waste disposal, etc., were not estimated separately since they were considered to be a part of the normal solvent extraction costs, since the costs should be nearly the same as those for standard, metal-clad oxide fuel on an equivalent throughput basis.

TABLE 3. — Capital-Cost Estimate for HTGR Head-End Facility

Item	Cost *
Building and services	$ 550,000
Building equipment	130,000
Cell structure	1,295,000
Cell services	565,000
Cell equipment	247,000
Process equipment	1,098,000
Process piping	906,000
Process and radiation instrumentation	350,000
Outside equipment	346,000
Site improvements	50,000
Utilities	85,000
Subtotal	$5,622,000
Engineering and inspection (20 %)	$1,124,000
Subtotal	$6,746,000
Contingency (25 %)	$1,687,000
Subtotal	$8,433,000
Interest during construction, startup costs, and working capital	$ 607,000
TOTAL	$9,040,000

* Installed cost, including contractor's overhead and profit.

The translation of these cost estimates into unit costs, per kilogram of thorium plus uranium, or per kwhr (electrical), depends critically on the actual fuel load (compared with the nominal capacity) and on the limitations imposed by the associated solvent extraction plant. The head-end facility might be built at a large multipurpose processing plant of the future, with a solvent extraction processing rate of several metric tons a day. Thus, the burn-leach lines might operate full-time, accumulating adjusted feed in the surge tank for a few solvent extraction campaigns per year. It might be built as a part of an integrated single-purpose HTGR fuel-processing plant with solvent-extraction capacity matched to the burn-leach capacity. If such a head-end facility were built in the next several years, it might be located at a relatively small multipurpose plant, such as the NFS plant, where the fraction of solvent extraction capacity that could be allocated to HTGR elements would limit the

HTGR head-end facility to part-time operation. In any of these three instances, if this head-end facility were built in time to process the first fuel discharged from the first HTGR, the processing load initially would represent only a fraction of nominal capacity, and it would be several years before capacity was reached, depending on the growth rate of the HTGR industry. The same problems in calculating processing costs were originally faced for estimating the cost of processing elements from the first commercial power reactors. Costs were determined on an interim basis by a USAEC-guaranteed reprocessing charge [15], based on cost estimates for a conceptual [16] "Reference Fuel-Processing Plant", and then by the NFS-USAEC contract [17]. This contract provided for charges based on a fully loaded plant, with the government providing a base load to partially make up for the lack of adequate private load during the early growth period of the power reactor industry. There is at present no equivalent accepted basis for determining charges for HTGR fuels; so the discussion which follows is highly speculative.

The NFS plant has a nominal capacity of 1,000 kg of uranium per day for 225 days a year. This is for standard uranium fuels of up to 3 % enrichement and burnups of 20,000 Mwd/metric ton. For higher enrichments and burnups, and for small fuel batch sizes, the effective processing rate may have to be reduced. For thorium fuels, the nominal capacity is 500 kg of thorium plus uranium per day. The reported capital cost is about $30,000,000, including site, fuel receipt and storage, chop-leach head-end, solvent extraction, waste disposal, engineering and interest, working capital and startup. The nominal annual charges are $7,050,000 including both capital and operating costs; this corresponds to about 0.20 mill/kwhr (elcctrical) if all the fuel were from typical, large, pressurized- or boiling-water reactors.

Based on a superficial comparison of building and equipement sizes, and on the HTGR head-end facility cost estimate described above, we see no reason why an integrated, single purpose processing plant for HTGR fuel, consisting of our conceptual head-end facility plus matching solvent extraction and other facilities, should have capital or operating costs significantly greater than those for the NFS plant, except that the costs for oxygen and alumina would be extra. Assuming NFS-type financing, such a reference plant could charge $75 to $80 per kg of uranium plus thorium, equivalent to a power-cost component of 0.11 to 0.15 mill/kwhr (electrical), depending on burnup and thermal efficiency. This is an attractively low prospective cost, but there may not be enough HTGR's to fully load such a plant until 1980 to 1985.

A much more conservative approach would be to assume that our conceptual HTGR head-end facility would be built at NFS in 1973, that it would have a load increasing from 3 metric tons a year in 1973 to 51 in 1980, that the HTGR head-end capital investment must be recovered, with interest, profit, and taxes by 1980 (the end of the nominal 15-year plant life for the NFS facilities), and that HTGR fuel processing costs must pay the full, regular, NFS charges in addition to the special head-end charges. A present-worth economic analysis [18], using a discount factor

of 8 to 10 % and a corporation income tax rate of 50 %, of such a hypothetical case indicated the following :

Special head-end capital and operating charges	\$107-134/kg(Th + U)
Regular NFS charges for thorium fuels	\$56-79
Total	\$163-213/kg(Th + U)

This total corresponds to 0.2 to 0.4 mill/kwhr (electrical) and is high enough to be a barrier to consideration of this approach to commercial HTGR fuel processing. On the other hand, since the total is based on a fairly pessimistic set of assumptions, extending the economic plant life plus providing some kind of base-load guarantee would permit substantially lower charges.

A significant technical assumption that would permit lower charges in a small HTGR fuel processing plant would be to specify a fuel element from which the fuel particles can be poured into a shipping container at the reactor. This would reduce shipping costs and permit cheaper fuel receipt and storage facilities at a processing site or the use of existing facilities at the NFS plant. In addition, the burner could be much smaller, with a single burner line possibly fitted into present cell space in the NFS plant.

7. — Combustion-Fluorination Process

Burning of graphite fuels in fluidized beds of inert alumina, followed by fluorination of the ash, is a potential nonaqueous alternative to the burn-leach method. Preliminary experiments [3] at Brookhaven National Laboratory indicate that it might be possible to achieve acceptable uranium recoveries from fuel that contains carbide particles. Pilot plant oxidation-fluorination experiments with prototype Peach Bottom fuel were conducted in a 4-in. -diam fluidized bed. The fuel was burned at 725 to 800° C in fluidized Norton R. R. alumina (equal parts of 60-, 90-, and 120-mesh). The product bed contained about 14 % ThO_2, 3 % U_3O_8, and 83 % Al_2O_3. After combustion, the bed was fluorinated at 450 to 550° C with F_2-N_2 mixtures containing between 25 and 50 vol % F_2. Uranium recoveries (as UF_6) were 92.1 and 98.8 % after fluorination times of 2.4 and 3.8 hr. A very exothermic reaction occuring above 500° C was controlled only by reducing the fluorine concentration in the fluidizing gas. However, since little UF_6 evolved at temperatures below 500° C, operation in the range 500 to 550° C appears mandatory if acceptable uranium recoveries are to be achieved. One other limitation of the combustion-fluorination process was noted in the preliminary tests. The thorium fluoride concentration in the bed must be held below 38 wt % to keep the bed from caking. More experiments

will be required to determine whether or not acceptable uranium recoveries from fuel that contains carbide particles can be achieved by the combustion-fluorination process. Even if such conditions are found, loss of thorium to the bed seems inevitable because of the extremely low volatility of ThF_4.

Preliminary experiments [19] indicate that the combustion-fluorination method will not be applicable to fuels that contain high-density ThO_2-UO_2 microspheres especially if the microspheres do not crack or break during irradiation and combustion. The reactivity of 150- to 180-μ-diam 96 % ThO_2 — 4 % UO_2 sol-gel microspheres in fluorine was tested on a laboratory scale. In a 4-hr experiments at 480° C, the microspheres were unaffected. Also, at 650° C, only 15 % of the uranium was volatilized as UF_6 in 4 hr; this was equivalent to a radial penetration of only 6 μ.

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REFABRICATION TECHNOLOGY AND COSTS FOR HIGH-TEMPERATURE GAS-COOLED REACTOR FUELS (1)

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ABSTRACT

High-temperature gas-cooled reactors utilizing the thorium-uranium-233 cycle have the potential of achieving low power costs and high fuel utilization. However, the realization of this potential depends on the development of technology for the economical refabrication of the fuel of these reactors. This paper describes the findings of the Oak Ridge National Laboratory in its investigation of this problem.

A brief description of typical nuclear fuels for HTGR reactors is followed by a discussion of the many problems concerning the refabrication of a hypothetical fuel element. Alternate fabrication flowsheets are shown, and data are presented which describe the effect of the amount of radioactive material in the fuel on fabrication plant design. Fuel refabrication costs are discussed in the second part of the paper. The effect of U^{232} concentration, mode of fabrication, amortization rate, plant capacity, and alternative fuel designs are represented by tables and curves. The third part of the paper describes the fuel refabrication program of ORNL. This includes a description of the ORNL Thorium-Uranium Recycle Facility (TURF) and the plans for equipment which is to be used in the facility for demonstration of refabrication technology. The investigation indicates that several refabrication process improvements are in sight at ORNL and that the cost penalties associated with refabrication of HTGR fuels are not prohibitive.

1. — INTRODUCTION

High-temperature gas-cooled reactors using the thorium-uranium-233 cycle are under development in the United States because of their potential for achieving low power costs and high fuel utilization. The attainment of low power costs depends to a great extent upon the development of technology for the economical recycle of fuel from such reactors, particularly coated-particle fuels. [1] Technical development is required for shipping, chemical processing, fuel preparation, and refabrication of bred fuel. Since the cost of fabrication of fuel is a major contributor to the cost of any fuel cycle, it is imperative that we consider the technology and the economics that are involved in refabrication of high-temperature gas-cooled reactor fuels.

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(1) ORNL-report TM-1115, May 1965.

(2) Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation (No. W.7405-eng-26).

The task of assessing the refabrication technology and the costs for refabricating HTGR fuel is difficult for two reasons. First, current fuel element designs are based principally on processes optimized for initial and, therefore, contact fabrication. This could lead to heavy penalties in calculating refabrication costs because certain contact steps become extremely awkward and expensive when they are automated and performed under the conditions imposed by recycle fuel. If we assume a design and a fabrication technique suitable for refabrication, we may err in estimating certain costs which are sensitive to assumptions regarding manufacturing tolerances. Second, there is a lack of experience in several key areas : (1) fabrication of fueled graphite fuel elements has not been done on a significant scale; (2) there is little experience with recycle fuel and none with refabrication of fueled graphite; and (3) there is no relevant experience with large plant processing or the economies to be effected in large plants fabricating fuel elements for power reactors.

At the Oak Ridge National Laboratory, we have taken some preliminary steps to an understanding of areas of technology in which information is incomplete. We are engaged in the ORNL Thorium Utilization Program, which has as its prime objective the eventual economical utilization of thorium in power reactor systems. The objectives of the program include the refabrication technology for various fuel elements; among those being studied are fuel element design concepts for high-temperature gas-cooled reactors. We are attempting to develop suitable technology for economical recycle of fuel in such reactors.

In developing a technology for economical recycle of fuel, we are placing a strong emphasis on the economics of refabrication. Therefore, we have done extensive cost analyses to determine areas where work should be performed; that is, to delineate those areas that appear most promising for a return on the investment of the development dollar. We have also performed many cost analyses to evaluate reactor systems and to compare the high-temperature gas-cooled reactors with other competitive reactor systems [2].

We are concerned in this paper with the speculative refabrication technology for fuel elements that would be used in high-temperature gas-cooled reactors. The text will discuss our preliminary plans for the refabrication of HTGR fuel and our economic analyses to determine the incremental costs in the process. We shall project the cost of fabricating one such fuel element using remote fabrication technology and shall offer comparisons of the cost of fuel refabrication with the cost of fabrication of the original U^{235}-bearing cores.

2. — Fuels for High-Temperature Gas-Cooled Reactors

Table 1 lists the main characteristics of the fuels of typical high-temperature gas-cooled reactors (i.e., AVR, Dragon, Peach Bottom, and an advanced HTGR concept). These fuels have a common characteristic — they contain carbides of uranium, thorium, or zirconium, or of a mixture of those elements. These carbides are in the form of microspheres or of nearly spherical particles (150 to 1,000 μ in diame-

TABLE 1. — Typical Fuel Elements for High-Temperature Gas-Cooled Reactors.

Reactor Name	Core Zone	Fertile Material	Other Materials	Fuel Microform			Fuel Macroform				Fuel Element			Fuel Elements per Bundle	References
				Kernel		Coating Material	Geometry	Dimension (in.)			Form	Dimension (in.)			
				Compounds	Geometry			OD	ID	H		OD	L		
AVR		U^{235} 93 % enrichment	Th Th/U = 8	Carbides	150-μ microspheres	80-μ pyrolytic carbon	No intermediate shape				Graphite sphere	2.4		1	[3—4]
Dragon	A	U^{235} 93 % enrichment	Th Th/U = 15	Carbides	250-420-μ microspheres	100-μ SiC + pyrolytic carbon	Annual compacts	1.75	1.37	2.0	Graphite tube	2.88	90	7	[5—8]
	B	U^{235} 93 % enrichment	Zr Zr/U = 5	Carbides	250-420-μ microspheres	100-μ pyrolytic carbon	Annular compacts	1.75	1.37	2.0	Graphite tube	2.88	90	7	[6—9]
Peach Bottom		U^{235} 93 % enrichment	Th Th/U = 8	Carbides	100-400-μ microspheres	50-60-μ pyrolytic carbon	Annular compacts	2.7	1.7	1.5	Graphite tube	3.5	144	1	[9—12]
Advanced HTGR Concept		U^{235} 93 % enrichment	Th Th/U = 27	Carbides	UC_2 : 200-μ microspheres ThC_2 : 1,000-μ microspheres	100-μ pyrolytic carbon 100-μ pyrolytic carbon	None (loose particles in 14 cylindrical cavities in each graphite rod)				Graphite tube	4.5	240	1	[2]

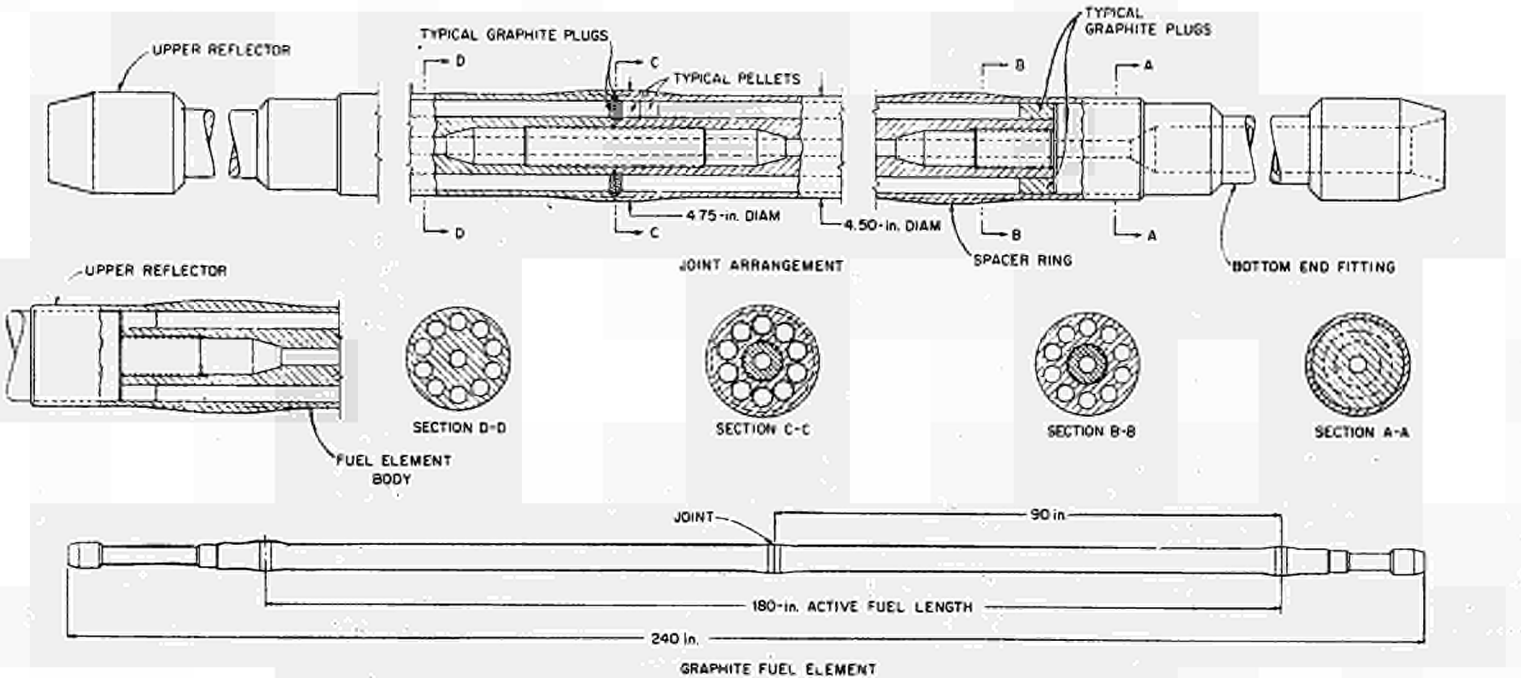


FIG. 1. — A Reference Design for an HTGR Fuel Element for Remote Fabrication.

ter). These particles are covered with an impervious fission-product-retaining coating (50 to 100-μ thick) which consists of pyrolytic carbon, either alone or in combination with silicon carbide.

The methods of agglomeration of the coated particles to form fuel elements vary widely. General Atomic [1] has proposed for advanced HTGR concepts that the coated particles be poured loosely into cavities of the graphite fuel element bodies. In the AVR reactor, coated particles are dispersed in a graphite matrix within each spherical fuel element. In the Dragon and Peach Bottom reactors, the coated-fuel particles are blended with graphite powder and pressed into intermediate-size annular compacts; these compacts are then placed into graphite tubes to form the fuel elements.

Fabrication techniques reflect these characteristics of the fuels, and consequently, the various methods used for preparing carbides and for coating particles have many similarities. However, the processes used for introducing these particles into the fuel element matrix differ widely from one designer to another.

To make this paper meaningful, we must discuss the fabrication of a fuel element of a specific design. Therefore, we shall use as the reference fuel element in these analyses a hypothetical design which, if certain developments are successful, should be useful in high-temperature gas-cooled reactors. This fuel element, shown in Fig. 1, has a diameter of 4 1/2 in. and has 10 equally spaced 3/4-in.-diam holes in the cross section between the outside diameter and the 3/4-in.-inside diameter. These holes are filled with fuel bodies consisting of an aggregate of coated fuel particles held together by a graphite matrix. The fuel element is 20 ft in overall length, and has two identical fuel subassemblies, each having an active fuel length of 7 1/2 ft. These fuel subassemblies can be fabricated separately, can be attached to either a reflector assembly or a fission-product trap assembly, and can be joined by the central coupling to form a complete fuel element.

3. — Refabrication of HTGR Fuel

Although we have selected a specific fuel element design for fabrication, there are some major decisions to be made before the refabrication of HTGR fuel can be attempted. One must choose the method of making spherical particles, the exact flowsheet to be used in fabrication, and the expected isotopic content of the processed fuel. We have studied a number of alternatives to be discussed subsequently, which must be considered in the refabrication of virtually any fuel element for high-temperature gas-cooled reactors.

3.1. — Preparation of Fuel Particles.

Heretofore, the technology for making spherical particles has consisted of various mechanical methods of consolidating combinations of the solid materials

(1) Division of General Dynamics.

of UO_2, ThO_2, and carbon and then treating these at high temperature to complete the necessary reactions and to densify the material. These methods invariably involve tedious techniques which become more complicated when special operating conditions are imposed in the recycle of fuel. Therefore, an objective of ORNL has been to simplify the manufacture of spherical particles.

We believe the sol-gel process [13] meets this objective. Clinton [14] at ORNL has demonstrated on a pilot scale the preparation of thoria-urania microspheres, which can be used as oxide or converted to carbide, as we shall discuss later. Following the flowsheet for the process, shown in Fig. 2, the microspheres are prepared by dispersing the sol at room temperature in an immiscible organic liquid which has some solubility for water. The sol droplets must be suspended in the organic phase until enough water is extracted from the aqueous sol to cause gelation. After drying, the gel spheres are calcined at 1150° C to complete the preparation of dense (99.4 % of theoretical is typical) oxide microspheres.

We shall assume that oxide microspheres, whether ThO_2, UO_2, or $(U\text{-}Th)O_2$, have been prepared by the sol-gel process.

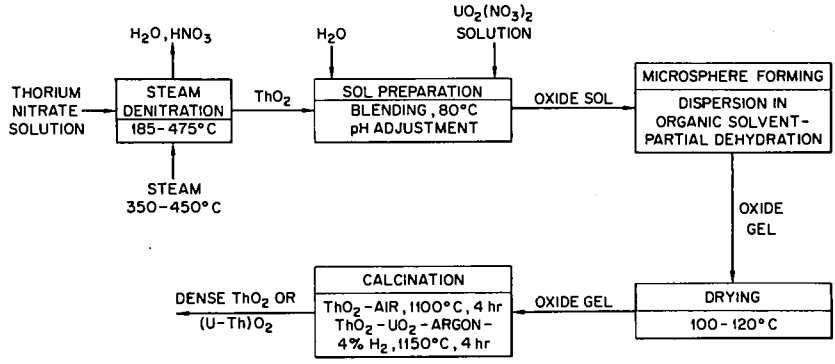


FIG. 2. — Flowsheet for Preparation of Oxide Microspheres by the Sol-Gel Process.

3.2. — FABRICATION FLOWSHEET.

Before discussing the details of a manufacturing flowsheet for large-scale production of HTGR fuel, we shall first consider the various alternatives which are possible in the fabrication of HTGR fuel. The alternatives are depicted in Fig. 3. One can use homogeneous fuel particles containing $(U\text{-}Th)C_2$ as a solid solution, or discrete particles of UC_2 and ThC_2. The use of discrete particles would allow the preparation of virgin ThC_2 in a hooded plant. There is also the possibility of using either mixed or discrete oxide particles instead of carbide. These alternatives are depicted as optional materials on the flowsheet.

To convey our impressions of how the manufacturing operations involved in the flowsheet would proceed, we shall describe briefly the process that starts with $(U\text{-}Th)O_2$ sol-gel microspheres. We envision that microspheres would be fed to a

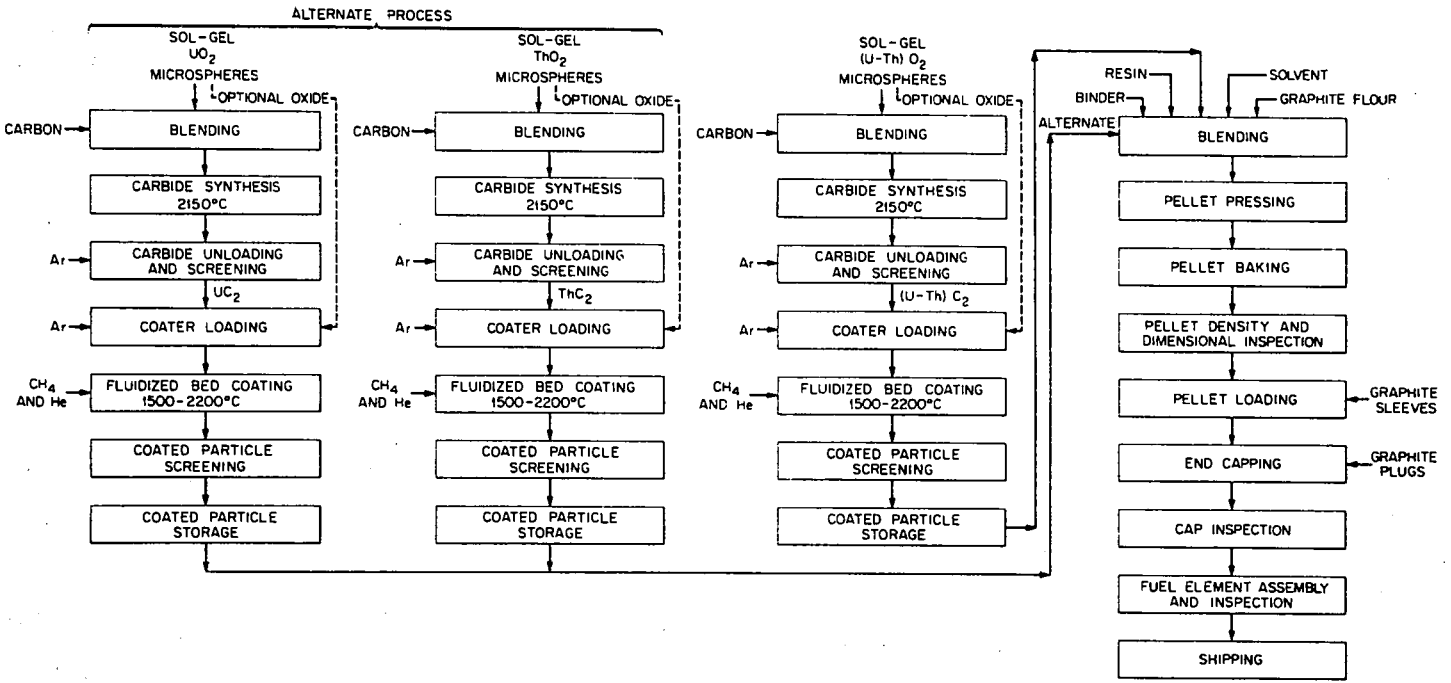


FIG. 3. — Flowsheet for Fabrication of HTGR Fuel Elements.

continuous rotating induction-heated graphite tube furnace along with carbon. As the particles move through the furnace, they would be reacted at 2,150° C to form carbide. On a continuous basis, the carbide-carbon mixture would be unloaded and the carbide separated from the carbon. The particles would be conveyed under inert atmosphere to the coaters, where the particles would be coated in batches at temperatures ranging from 1,500 to 2,200° C.

The particles would then be blended with resin, graphite flour, binder, and solvent; the mixture would be pressed into cylindrical compacts on automatic presses and subsequently baked in continuous furnaces at approximately 1,000° C. The compacts would be inspected for density and conformity to dimensional specifications.

The pellets would be loaded into the holes in the graphite segment; the holes capped with a graphite plug, sealed, and inspected. Finally, two segments would be joined by a threaded or bayonet central fitting and sealed. After inspection of the seal for leaks and the element for dimensions, weight, and transferrable contamination, the element would be shipped. In addition to these principal steps, routine inspections and tests would be performed to assure maintenance of quality.

3.3. — Effect of Fuel Isotopic Content on Fabrication Plant Design.

Several recycling systems can be used for HTGR fuel. For example, one may choose not to recycle the thorium, but to recycle the U^{233}. With (U-Th)C_2 particles, all of the material would be handled in the refabrication plant; but when discrete particles are used, the virgin thorium could be prepared in a hooded operation. The other alternative is to recycle the thorium and the U^{233}.

Thus, in any proposed fabrication plant, one has the problem of selecting for the given isotope the type or mode of fabrication that is to be employed in the fabrication of either first cycle or recycle fuel. The possibilities are contact, hooded, glove box, semiremote, or remote operation. We have chosen to define these terms as follows :

(1) Contact operations are those in which the operator has direct contact with the material.
(2) Hooded operations are those which are contained in ventilated enclosures that are not hermetically sealed.
(3) Glove box operations are those requiring hermetic sealing of the equipment.
(4) Semiremote operations are those requiring light shielding.
(5) Remote operations are those requiring heavy shielding and totally remote manipulation.

The type and quantity of the isotope in the fuel and the quantity of fuel being processed dictate the type of operation to be selected. In the fuel cycle of interest, thorium-uranium-233, the mode of fabrication depends upon the U^{232} concentration in the fuel, its age, and whether virgin or recycle thorium is being used. The amount of fuel being processed, its concentration, and its proximity to the operator is also important. We have analyzed the effect of U^{232} concentration on the type of facility which should be employed in the fabrication of HTGR fuel. In this analysis, we used the flowsheet shown in Fig. 3 and considered both discrete particles of UC_2 and ThC_2

TABLE 2. — Material Location, Quantity, and Age in Fabrication Plant Processing (U-Th)C_2

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
Zone 1				
(1) In-process material at a given time, kg	22	88	352	1,408
(2) Time since in-process material received from sol-gel plant, hr	16	16	16	16
(3) Material held up in equipment, kg	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	5.7	5.7	5.7	5.7
Zone 2				
(1) In-process material at a given time, kg	20	80	320	1,280
(2) Time since in-process material received from sol-gel plant, hr	26	26	26	26
(3) Material held up in equipment, kg	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	6	6	6	6
Zone 3				
(1) In-process material at a given time, kg	15	30	60	60
(2) Time since in-process material received from sol-gel plant, days	1.7	1.3	1.3	1.2
(3) Hold-up material in any element, kg.	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	6.7	6.4	6.3	6.3

and particles of (U-Th)C_2. To perform the shielding calculations and to determine the mode of fabrication, the fuel element fabrication facility was divided into three zones.

Zone 1. The oxide microspheres are received from the sol-gel facility, converted to carbide, and inspected.

Zone 2. The fuel particles are coated with pyrolytic carbon and inspected.

Zone 3. Compacts are prepared, inspected, and loaded into the graphite sleeves. Finally, two fuel segments are assembled together with end pieces, inspected, and shipped to the reactor site.

Shielding was calculated for plants having daily processing capacities ranging from 10 to 3,700 kg of heavy metal per day. Typical material flow rates for (U-Th)C_2 particles are shown in Table 2; those for UC_2 particles in Table 3.

The following assumptions were made in calculating shielding requirements :

1. The time between solvent extraction and receipt of material at the fuel element fabrication plant is five days.
2. A major cleanup of the equipment and enclosures is performed after five working days.

TABLE 3. — HTGR Material Location, Quantity, and Age in Fabrication Plant Processing UC_2 and ThC_2.

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
Zone 1				
(1) In-process material at a given time, kg	3	4	16	64
(2) Time since in-process material received from sol-gel plant, hr	16	16	16	16
(3) Material held up in equipment, kg	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	5.7	5.7	5.7	5.7
Zone 2				
(1) In-process material at a given time, kg	3	6	24	96
(2) Time since in-process material received from sol-gel plant, hr	26	26	26	26
(3) Material held up in equipment, kg	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	6	6	6	6
Zone 3				
(1) In-process material at a given time, kg	15	30	60	60
(2) Time since in-process material received from sol-gel plant, days	1.7	1.3	1.3	1.2
(3) Hold-up material in any element, kg	3	3	3	3
(4) Time since hold-up material received from sol-gel plant, days	6.7	6.4	6.3	6.2

3. No substantial quantity of material is located closer than 1 ft to the enclosure wall.
4. The amount of material retained in the equipment during processing is 3 kg, but the material is released and continued in process at five-day cleanup intervals.
5. The plant processes 110 % of the quantity shipped.

The results of our shielding calculations for semiremote and remote plants fabricating HTGR fuel elements with $(U\text{-}Th)C_2$ particles are shown in Table 4. The results for fuel elements containing discrete particles of UC_2 and ThC_2 are shown in Table 5. As can be observed in the tables, we have used approximately 3.5 in. of steel as the practical limit for semiremote fabrication because of the difficulty of working through a greater distance with gloved hands or tongs. Also, radiation from sources requiring greater than 3.5 in. of steel would prohibit, or at least greatly inhibit, contact maintenance of equipment. It should be noted that there are differences in shielding requirements for fabrication of $(U\text{-}Th)C_2$ particles and fabrication of discrete UC_2 and ThC_2 particles. The discrete particles of UC_2 require greater shielding because

TABLE 4. — Shielding for Semiremote and Remote Plants Fabricating Fuel Elements Containing $(U\text{-}Th)C_2$ Particles

ppm [a]	Plant Capacity (kg heavy metal/operating day)									
	10	60	100	230	500	750	930	1,250	1,500	3,700
	Steel (in.)									
1	0.2	0.2	0.6	1.2	1.6	1.8	1.9	1.9	1.9	2.0
2	1.1	1.1	1.4	2.0	2.4	2.6	2.7	2.8	2.8	2.9
5	2.2	2.2	2.5	3.1	3.5					
10	3.0	3.0	3.4							
	Normal Concrete (in.)									
5						12.4	12.8	13.0	13.2	13.9
10				13.2	14.5	15.3	15.6	15.9	16.1	17.0
20	12.7	12.7	14.0	16.0	17.3	18.1	18.4	18.8	19.0	20.0
50	16.4	16.4	17.7	19.6	21.0	21.8	22.2	22.6	22.9	24.1
100	19.2	19.2	20.4	22.4	23.9	24.6	25.0	25.5	25.8	27.2
500	25.6	25.6	26.9	28.8	30.4	31.2	31.6	32.2	32.5	34.3
1,000	28.4	28.4	29.6	31.6	33.2	34.0	34.4	35.1	35.4	37.4
Recycle Th		33.0		36.0			39.0			42.5

[a] Parts per million U^{232} in heavy metal.

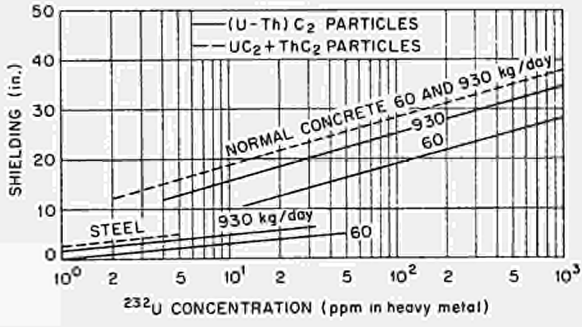
TABLE 5. — Shielding for Semiremote and Remote Plants Fabricating Fuel Elements Containing UC_2 and ThC_2 Particles

ppm [a]	Plant Capacity (kg heavy metal/operating day)									
	10	60	100	230	500	750	930	1,250	1,500	3,700
	Steel (in.)									
1	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.9	3.0	3.2
	Normal Concrete (in.)									
2	12.3	12.3	12.3	12.3	12.3	12.3	12.3	12.5	12.6	13.5
5	15.9	15.9	15.9	15.9	15.9	15.9	15.9	16.1	16.3	17.2
10	18.7	18.7	18.7	18.7	18.7	18.7	18.7	18.9	19.1	20.0
20	21.4	21.4	21.4	21.4	21.4	21.4	21.4	21.7	21.9	22.8
50	25.1	25.1	25.1	25.1	25.1	25.1	25.1	25.3	25.5	26.5
100	27.9	27.9	27.9	27.9	27.9	27.9	27.9	28.1	28.3	29.3
500	34.3	34.3	34.3	34.3	34.3	34.3	34.3	34.5	34.7	35.8
1,000	37.0	37.0	37.0	37.0	37.0	37.0	37.0	37.3	37.5	38.6
Recycle Th		41.5		41.5			41.5			43.0

[a] Parts per million U^{232} in heavy metal.

they are not diluted with thorium during conversion and coating operations. In all of these results, the shielding is calculated to limit body exposure to 1 mr/hr.

The shielding required for 60 to 930 kg/day plants is plotted versus U^{232} concentration in Fig. 4. As can be seen, the limit for semiremote fabrication of UC_2 plus ThC_2 particles is approximately 2 ppm U^{232} in total heavy metal. Beyond 2 ppm for these particles, a designer would probably use normal concrete. The concentration of U^{232} limiting semiremote fabrication of $(U\text{-}Th)C_2$ is higher than that for discrete particles. The shielding requirements for fabrication of fuel elements containing $(U\text{-}Th)C_2$ particles vary with the amount of material being processed in the plant; as can be seen in Fig. 4, the 930 kg/day plant requires slightly more shielding than the 60 kg/day plant.



Shielding Required to Limit Body Exposure to 1 mr/hr.

FIG. 4. — Effect of U^{232} Concentration on Fabrication Plant Shielding.

Although we are not certain as to the U^{232} concentration that can be expected in equilibrium cycle fuel, 50 ppm in heavy metal would probably be a realistic value. We, therefore, conclude from our analysis that a remote plant is required for refabrication of such fuel.

4. — REFABRICATION COSTS

Several detailed cost studies for medium-temperature gas-cooled reactors have recently been published in the United Kingdon [15] and in France [16]; in the U. S. A., a few broad cost estimates have been presented for high-temperature gas-cooled reactors [2, 17, 18]. At the Oak Ridge National Laboratory, we have as an objective the complete analysis of the refabrication costs for any reactor fuel element, including fueled graphite elements. This objective requires a tremendous number of calculations. Therefore, during the past two years, we have developed a computer program for the calculation of the cost of fabricating fueled graphite elements. We have used this computer program to derive the data which will be presented subsequently.

4.1. — METHOD OF ANALYSIS.

The computer program was written from data evolved in the following manner. The basic equipment required in the plant was selected, and the uninstalled cost of this equipment was estimated. These costs were multiplied by various factors to determine the final capital cost of the fabrication plant. The multiplication factors were obtained from studies conducted jointly with construction engineers in which detailed plant layouts were examined for various sizes and types of fabrication plants. These studies yielded detailed breakdowns of such costs elements as building construction, equipment installation, instrumentation, engineering, and pre-operation charges. In the computer program, equations are included for each step in the process such that, upon description of the size and type of plant in which the fabrication is to occur, proper capital cost of that step can be obtained.

Operating costs were determined by similar methods. Material costs in the manufactured product were obtained through consultations with industrial manufacturers. In this manner, the costs, shown in Table 6, for the large pieces were obtained.

TABLE 6. — Basic Costs of Graphite Hardware.

Plant production rate, kilogram of heavy metal per operating day	10	100	500	1,000
Dollars per fuel element	300	290	270	230

Cost of Preparing Spherical Oxide Particles.

The computer program does not calculate the cost of preparing spherical particles of oxide which is the starting material in the fabrication flowsheet. But, the cost of producing spherical oxide particles by the sol-gel process has been estimated by Harrington and Chandler [19] and is shown in Fig. 5 for a large range of production rate and for processing the different isotopes of interest in the thorium fuel cycle. The cost of oxide particle preparation is not included, unless specifically noted, in the fabrication costs presented in this paper.

4.2. — BASIS FOR CALCULATION OF FABRICATION COSTS.

In calculating the fabrication costs which are subsequently presented, we used as a basis the fuel element shown in Fig. 1 and the flowsheet presented in Fig. 3. We made the following assumptions.

1. The fabrication plants would be single purpose; that is, only fueled graphite elements would be fabricated.

2. The (U-Th)C_2 particles would be 1,000 μ in diameter with 100 μ pyrolytic-carbon coating.

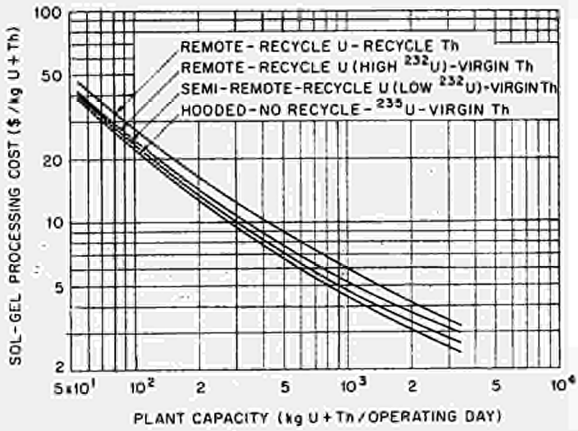


FIG. 5. — Cost of Preparation of Spherical (U-Th)O_2 Particles by the Sol-Gel Process.

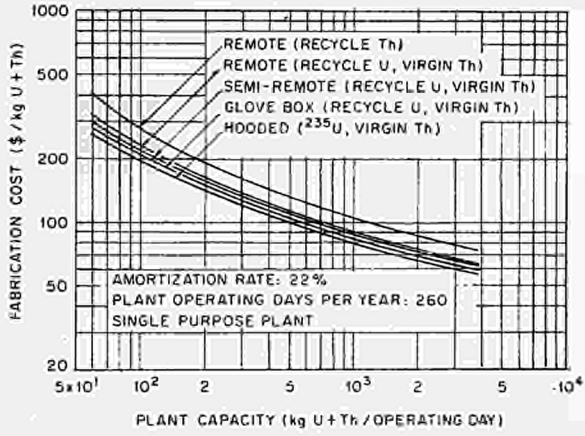


Fig. 6. — Effect of Mode of Fabrication on Costs of HTGR Fuel Elements Containing (U-Th)C_2 Particles.

3. When discrete particles of UC_2 and ThC_2 are used, the UC_2 would be 200 μ in diameter; the ThC_2 would be 1,000 μ in diameter; and both would have 100 μ of carbon coating.

4. The fabrication plants would operate 260 days per year, three shifts per day The data do not include charges for profit and for inventory or cost of source or fissionable material. Non-nuclear hardware costs are included.

4.3. — Effect of Mode of Fabrication and Production Rate on Cost.

One comparison of interest is that between the costs of various modes of fabrication. The cost of fabrication of fuel elements containing particles of (U-Th)C_2 in plants amortized at a rate of 22 % is shown in Fig. 6. The cost of fabrication of elements containing discrete particles of uranium carbide and thorium carbide amortized

at an annual rate of 22 % is shown in Fig. 7. Both figures present the cost of recycling of thorium; as can be seen, the penalty for recycling thorium as discrete particles is quite high. All of these curves show the substantial effect of production rate on fabrication cost. The predicted cost reductions are a factor of 4 when production rates are scaled from 60 to 3,700 kg/day.

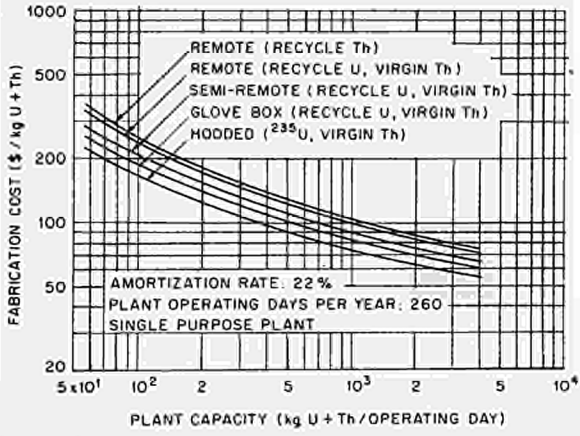


FIG. 7. — Effect of Mode of Fabrication on Costs of HTGR Fuel Elements Containing UC_2 and ThC_2 Particles.

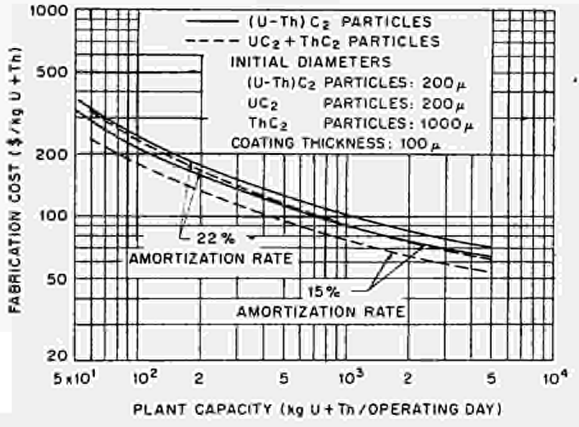


FIG. 8. — Effect of Amortization Rate and Type of Fuel Particles on Cost of Remote Fabrication.

4.4. — EFFECT OF RECYCLE METHOD AND AMORTIZATION RATE ON COST.

In Fig. 8 we have replotted the data to show the difference in cost of fabricating HTGR fuel elements containing recycle U^{233}, using $(U\text{-}Th)C_2$ particles in one instance and discrete particles of UC_2 and ThC_2 in a second case. The thorium in each case is virgin. The figure also compares amortization rates of 15 and 22 %. The curves indicate that there is little difference between the costs of fabricating the two fueling

systems, even though in the case of the discrete particles a substantial quantity of fuel is not being fabricated in a remote plant. Presumably, the fact that two separate fabrication facilities must be built and operated offsets the advantages of performing the conversion and coating of thorium-bearing particles in a hooded plant.

4.5. — Comparison of Costs of Oxide with Carbide Fueled Elements.

Figure 9 exhibits the cost difference between fabrication of fueled graphite elements containing carbide particles and those containing oxide particles. The curves are for remote fabrication (24-in. concrete shielding) of particles at a plant amortization rate of 22 %. The curves reveal that the savings in the cost of fabrication by using oxide particles in the fuel element would range from approximately 15 % at low production rates to 5 % at high production rates. A simplified process for conversion of oxide to carbide has been used for these calculations. If conventional technology were used, we believe that the cost of conversion would be somewhat higher.

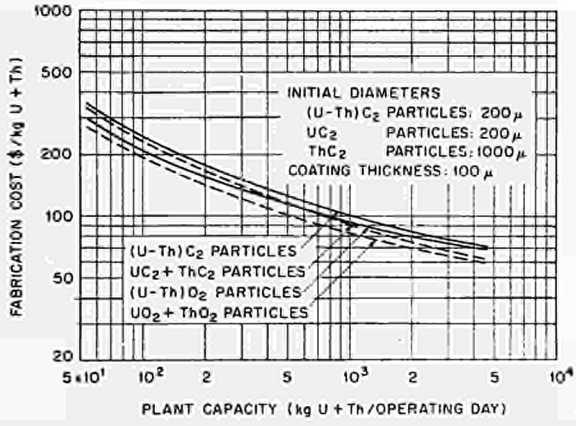


Fig. 9. — Comparison of Remote Fabrication Costs of Fuel Elements Containing Oxide and Carbide Particles.

4.6. — Variations in Cost as a Function of Shielding.

The effect of U^{232} concentration on the cost of fuel elements is presented in Fig. 10. Using plant amortization rates of 22 %, the effects are shown for both discrete particles and the particles of $(U\text{-}Th)C_2$. Curves are plotted for 60, 230, 930, and 3,700 kg/day plants. The curves indicate that as the plant capacity increases fabrication costs become more insensitive to the U^{232} concentration in the fuel. We do not know exactly the transition points between semiremote operation and remote operation; but, as can be seen from the curves, from the standpoint of cost it is not extremely important that the transition points be determined precisely.

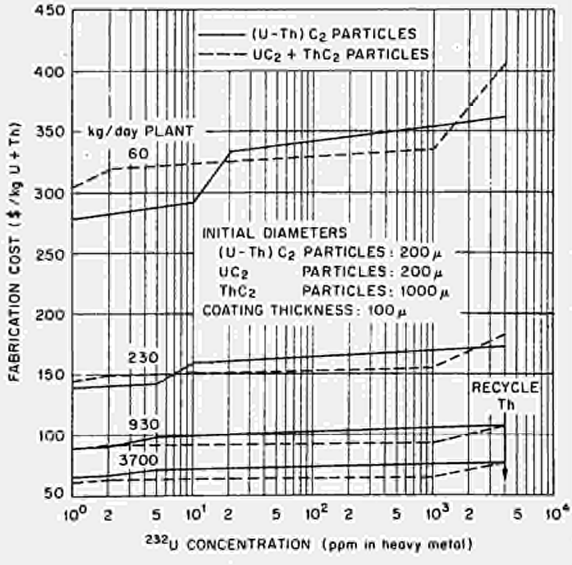


FIG. 10. — Effect of U^{232} Concentration on Cost of Refabrication.

4.7. — INCREMENTAL COST ELEMENTS.

Because of the importance of incremental cost elements, we have performed a detailed analysis of the factors which contribute to the cost for both hooded plants and remote refabrication plants. Tables 7 and 8, respectively, contain data for (U-Th)C_2 particles and discrete particles of UC_2 and ThC_2, both with concentrations of 50 ppm U^{232} in heavy metal.

The tables show that the capital cost per kilogram of fuel processed for conversion decreases rapidly with increasing plant capacity. The capital cost for assembly operations show the same trend. However, for the coating operations, the decrease in cost with increase in plant capacity is not comparable to the other two categories of operation. This is attributable to the fact that the coaters quite rapidly reach their maximum practicable size. Although some economy would be effected by purchasing multiple units, this is not sufficiently great to have a marked effect. Most of the effect of scale in the coating operation is caused by the peripheral equipment, such as weighing devices, conveyors, and inspection equipment, all of which are sensitive to production rate. In the assembly operation, one does not approach the maximum capacity of a single item of equipment at a very low production rate; and, therefore, the effect of plant capacity reflects the increased utilization of the equipment.

The operating costs shown in the tables reflect the effect of automation of the operation and the effect of a large amount of overhead in plants with very low production rates. Again, we observe that the production rate has less effect on coating cost than the other two categories of operation. Production rate also affects the hardware costs significantly.

TABLE 7. — Breakdown of Costs for Refabrication [a] with $(U\text{-}Th)C_2$ Particles.

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
	$/kg heavy metal			
Capital Cost				
Conversion	17	5	3	2
Coating	42	28	21	18
Assembly	74	28	14	8
Subtotal	133(71)	61(34)	38(22)	28(16)
Operating Cost				
Conversion	27	8	3	2
Coating	48	19	11	9
Assembly	97	43	23	15
Subtotal	172(117)	70(49)	37(27)	26(19)
Hardware Cost	33	32	27	19
TOTAL COST	338(221)	163(115)	102(76)	73(54)

[a] 50 ppm U^{232} in heavy metal; virgin thorium; 22 % amortization rate.
NOTE : Numbers in parentheses represent non-recycle fuel in hooded plant.

TABLE 8. — Breakdown of Costs for Refabrication [a] with UC_2 and ThC_2 Particles

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
	$/kg heavy metal			
Capital Cost				
Conversion	13	4	2	2
Coating	36	22	17	14
Assembly	74	28	14	8
Subtotal	123(95)	54(45)	33(28)	24(22)
Operating Cost				
Conversion	27	7	2	1
Coating	47	16	8	6
Assembly	97	43	23	15
Subtotal	171(136)	66(54)	33(28)	22(19)
Hardware Cost	33	32	27	19
TOTAL COST	327(264)	152(131)	93(83)	65(60)

[a] 50 ppm U^{232} in heavy metal; virgin thorium; 22 % amortization rate.
NOTE : Numbers in parentheses represent non-recycle fuel in hooded plant.

As can be seen in these tables, the coating operation accounts for approximately 25 % of the costs at a plant capacity of 60 kg/day in remote plants and 38 % of the costs in 3,700 kg/day plants. Obviously, in order to reduce costs, one should strive to increase the efficiency of the coating step.

Using the data in Tables 7 and 8, one can approximate the incremental cost factors which can be applied to similar fabrication processes to obtain the refabrication cost if the cost of fabricating non-recycle fuel is known. Tables 9 and 10 present the ratios of remote to hooded plant costs, which can be used for this purpose. It is clear that the capital and operating factors decrease as plant capacity increases. The decrease in these factors is understandable because there are certain costs which

TABLE 9. — Costs Ratios for Comparison of Remote and Hooded Plants Fabricating (U-Th)C_2 Particles [a]

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
	Ratio of Remote to Hooded Cost			
Capital	1.87	1.79	1.73	1.75
Operating	1.47	1.43	1.37	1.37
Total (including Hardware)	1.53	1.42	1.34	1.35

[a] 50 ppm U^{232} in heavy metal; virgin thorium.

TABLE 10. — Cost Ratios for Comparison of Remote and Hooded Plants Fabricating UC_2 and ThC_2 Particles [a]

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
	Ratio of Remote to Hooded Cost			
Capital	1.29	1.20	1.18	1.09
Operating	1.26	1.22	1.18	1.16
Total (including Hardware)	1.24	1.16	1.12	1.08

[a] 50 ppm U^{232} in heavy metal; virgin thorium.

are only first costs in remote operations. For example, health physics facilities are required in a 60-kg/day plant as well as a 3,700-kg/day plant; and probably the facilities would be of the same type and very nearly the same size in both cases. Thus, we are observing the effect of more efficient utilization of certain service personnel and facilities in the larger plants. It is notable that most of the difference falls between plant capacities of 60 and 230 kg/day, indicating that additional facilities and personnel are required for the next larger plant.

4.8. — Variations in Cost with Particle Size and Coating Thickness.

Another comparison which may be of interest is that of coating particles of different size and with different coating thicknesses. The effect of larger particles on coating efficiency is not accurately known; problems could be encountered in the decreased surface area per kilogram of fuel. A much increased gas velocity is required to fluidize the particles; there is poorer gas solid contact; the particle motion characteristics change; and there might be increased maintenance due to sooting.

From theoretical consideration of these effects, we derived the following relationship of initial particle diameter and coating efficiency :

$$CK = \left(\frac{D_0}{250}\right)^{0.1}$$

where

CK = coating efficiency coefficient,

D_o = initial particle diameter in microns.

This relationship was used in the computer program to adjust the cost of the coating operation. The factor was not applied to other processing steps. The fabrication costs at three production rates, 60, 230, and 930 kg/day, for fueled graphite elements

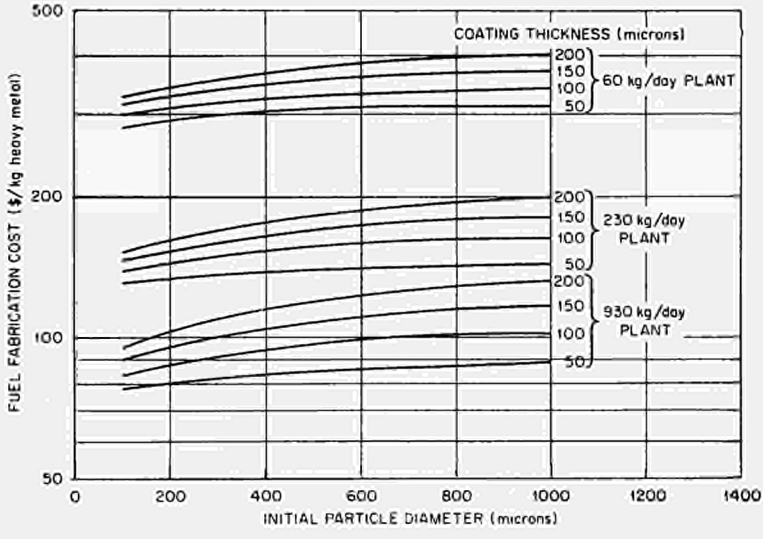


Fig. 11. — Relationship of Fuel Fabrication Cost to Particle Diameter and Coating Thickness for Elements Containing (U-Th)C_2 Particles.

containing particles of various diameters and various coating thickness are given in Fig. 11.

4.9. — Refabrication Cost of a Typical HTGR Fuel Element.

In the preceding discussion, we have been concerned with all of the factors which affect cost. As a final note to our discussion, we would like to present the predicted costs, including that for spherical particle preparation, for a typical fuel element that might be refabricated. We shall assume that the fuel element contains 8.8 kg of 400-μ diam $(U\text{-}Th)O_2$ particles with 100-μ pyrolytic carbon coating, that the fuel contains 50 ppm U^{232} in total heavy metal, and that the sol-gel and fabrication plant is amortized at an annual rate of 22 %. We believe the use of coated oxide particles to be a realistic projection of technology because of the excellent irradiation performance of this material to date [20]. The results are given in Table 11 for several production rates.

Table 11. — Fuel Preparation and Fabrication Costs for a Typical Recycle HTGR Element

	Plant Capacity (kg heavy metal/day)			
	60	230	930	3,700
	$/kg heavy metal			
Particle Preparation				
Capital Charge	14	5	1.4	1.3
Operating Charge	25	7	2.6	1.2
Fabrication				
Capital Charge	106	49	29	21
Operating Charge	135	57	31	21
Hardware	33	32	27	19
Total Cost	313	150	91	63.5

5. — ORNL Pilot-Scale Refabrication Program

We are conducting an extensive program for the development of an economical procedure for the remote, automated refabrication of HTGR fuel. Processes which are developed will be demonstrated in the Thorium-Uranium Recycle Facility (TURF), [21,22] which is now ready for construction at ORNL.

5.1. — Description of the Thorium-Uranium Recycle Facility (TURF).

The TURF will furnish the necessary space and shielding to perform all of the operations required for the processing of a spent HTGR fuel element through the

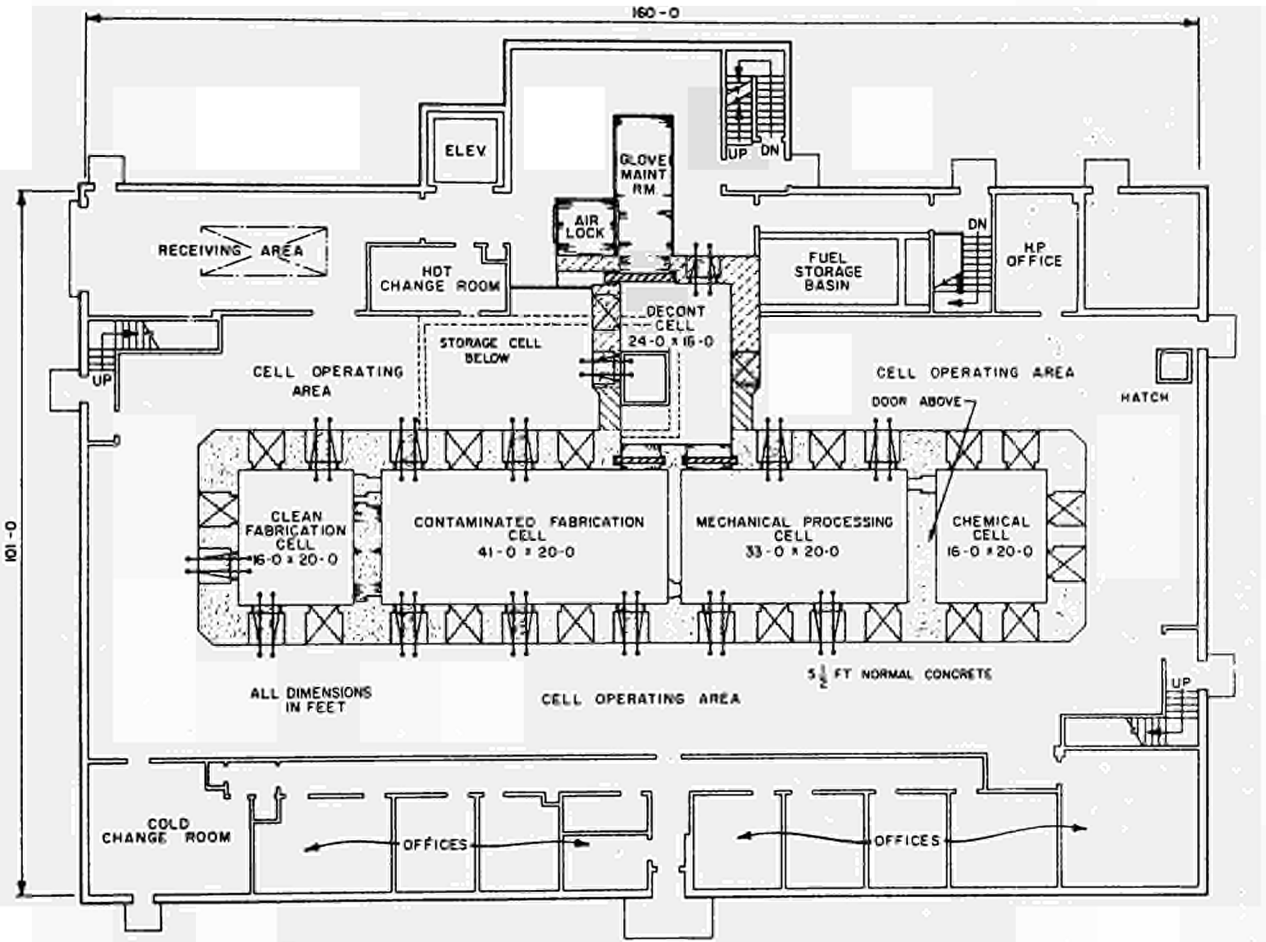


FIG. 12. — ORNL Thorium-Uranium Recycle Facility — First Floor.

various phases of the fuel cycle. The facility has been sized to accomodate integrated recycle processes with equipment scaled down from the anticipated production units so that a realistic and reliable basis for technical and economical analyses will exist.

The facility will be an irregularly shaped three-story building approximately 162 ft long, 124 ft wide, with a partial basement. The first floor, shown in Fig. 12, provides space for offices, change rooms, operating areas around the cell enclosures, a fuel storage basin, and a receiving area. The second floor provides space for chemical makeup, sampling of in-cell processes, cask decontamination, a shop for contamined equipment, a development laboratory, and the mechanical and electrical equipment for the building. The third floor is a high bay area which includes the cell roof area and provides facilities for entry of cell services and cell access. The bay is provided with a 50-ton crane to handle casks large enough to accomodate fuel elements up to 12 ft in length. The basement will provide space for access to the equipment storage cell and for the vulnerable equipment associated with the chemical cell. The building acts as a second line of containment for the cell complex.

The primary zone of containment for the facility consists of six shielded cells and associated glove maintenance room and air lock, all of which are depicted in Fig. 13. Four of the cells provide the operating space for the process equipment while two provide supporting functions. The mechanical processing and chemical processing cells will be used for operations incidental to irradiated fuel recovery and reconstitution of fissile and fertile materials into forms suitable for use in fuel element manufacture. The contaminated fabrication cell provides space for fabrication operations through the point where all fissile and fertile material is contained and sealed in fuel cladding. The clean fabrication cell will be used for final assembly and inspection of fuel elements. The two large cells, mechanical processing and contaminated fabrication, are to be maintained remotely; the clean fabrication cell will be maintained by a contact means. The chemical cell has the flexibility of allowing either method of maintenance.

All the remote maintenance cells and the decontamination cell are lined with stainless steel. The service penetrations for all cells are provided with seals to confine radioactivity. Provisions have been made for future conversion to an inert atmosphere in the remote maintenance cells, the decontamination cell, equipment storage cell, and the glove maintenance room to permit processing of pyrophoric materials on a large scale.

The cells are capable of processing and refabricating fuel assemblies as long as 12 ft and containing as much as 35 kg of Th-U fuel irradiated to 25,000 Mwd/MT and decayed for 90 days. All of the operating cells are provided with the equivalent of 5 1/2 ft of normal concrete up to the electromechanical manipulator bridge level and 4 1/2 ft of concrete above this level. Figure 14 is a section elevation showing the common roof line of all operating cells with the varying floor levels required to provide different in-cell height to permit processing and refabrication of power reactor size fuel elements. The section also shows the crane and electromechanical

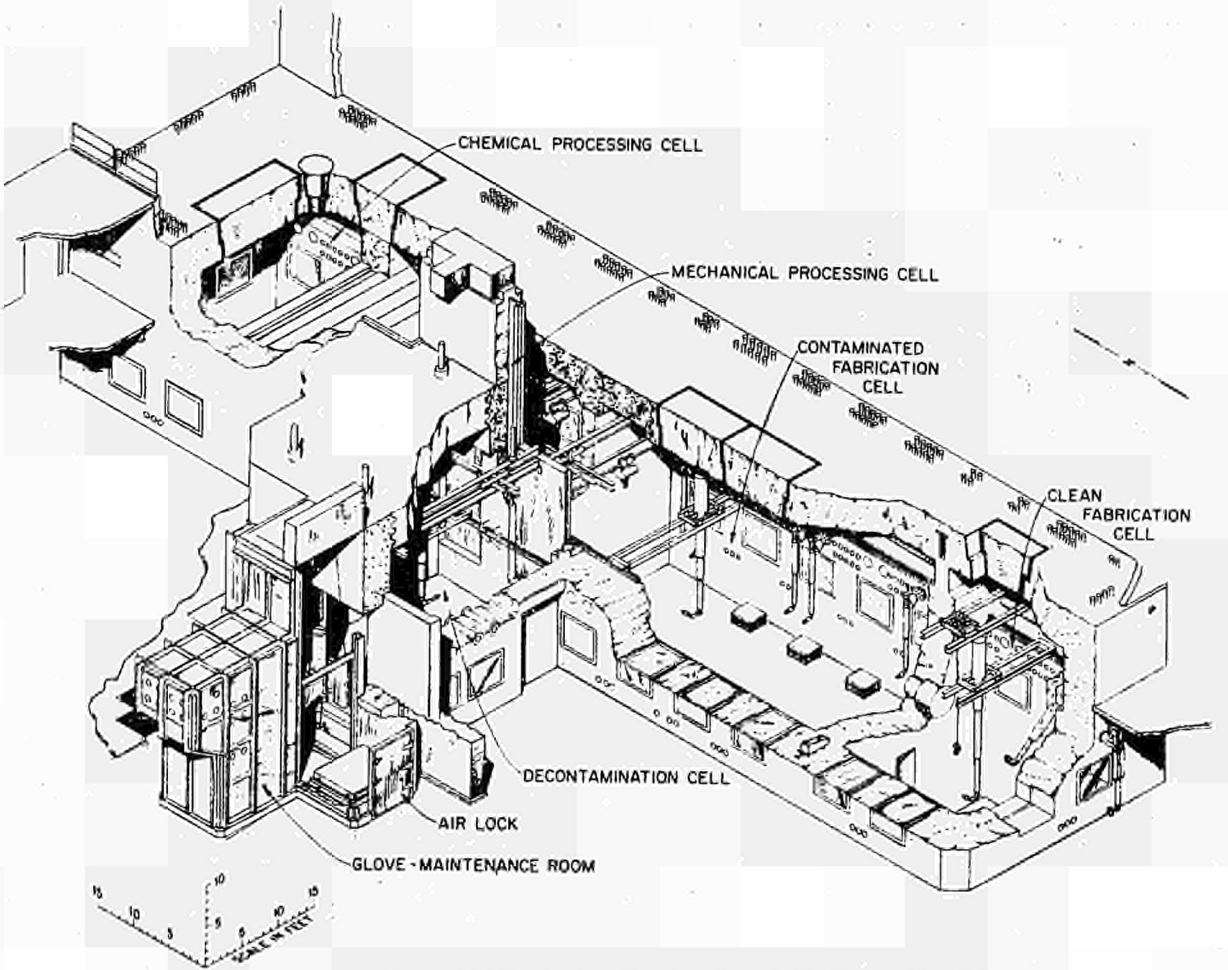


FIG. 13. — ORNL Thorium-Uranium Recycle Facility — Hot Cells.

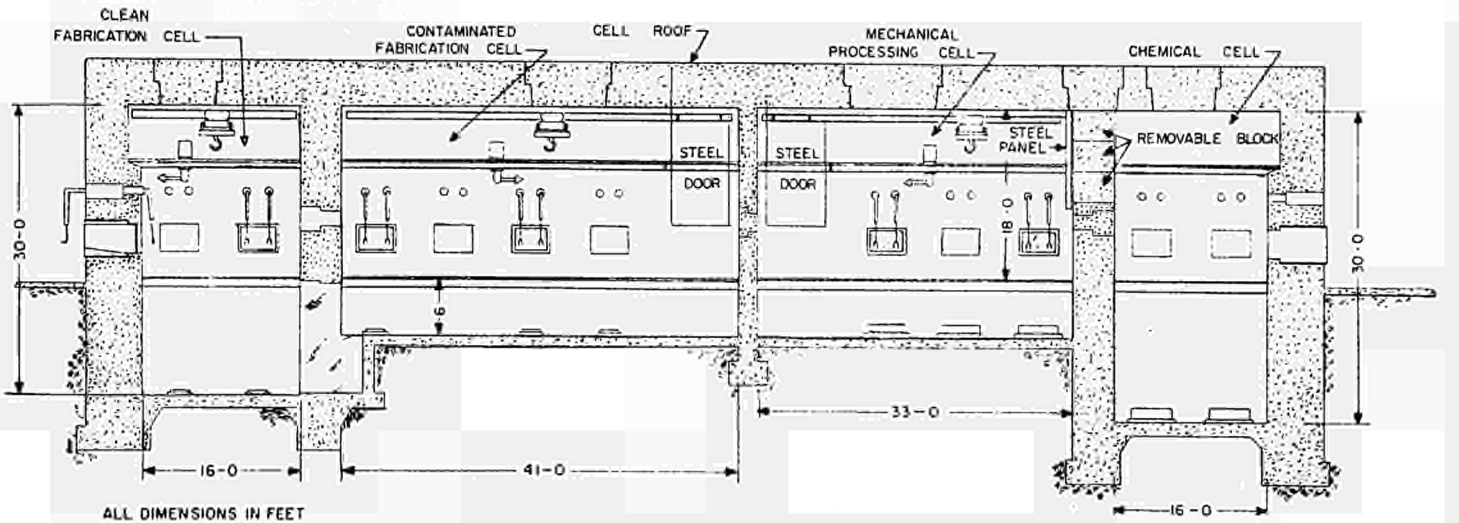
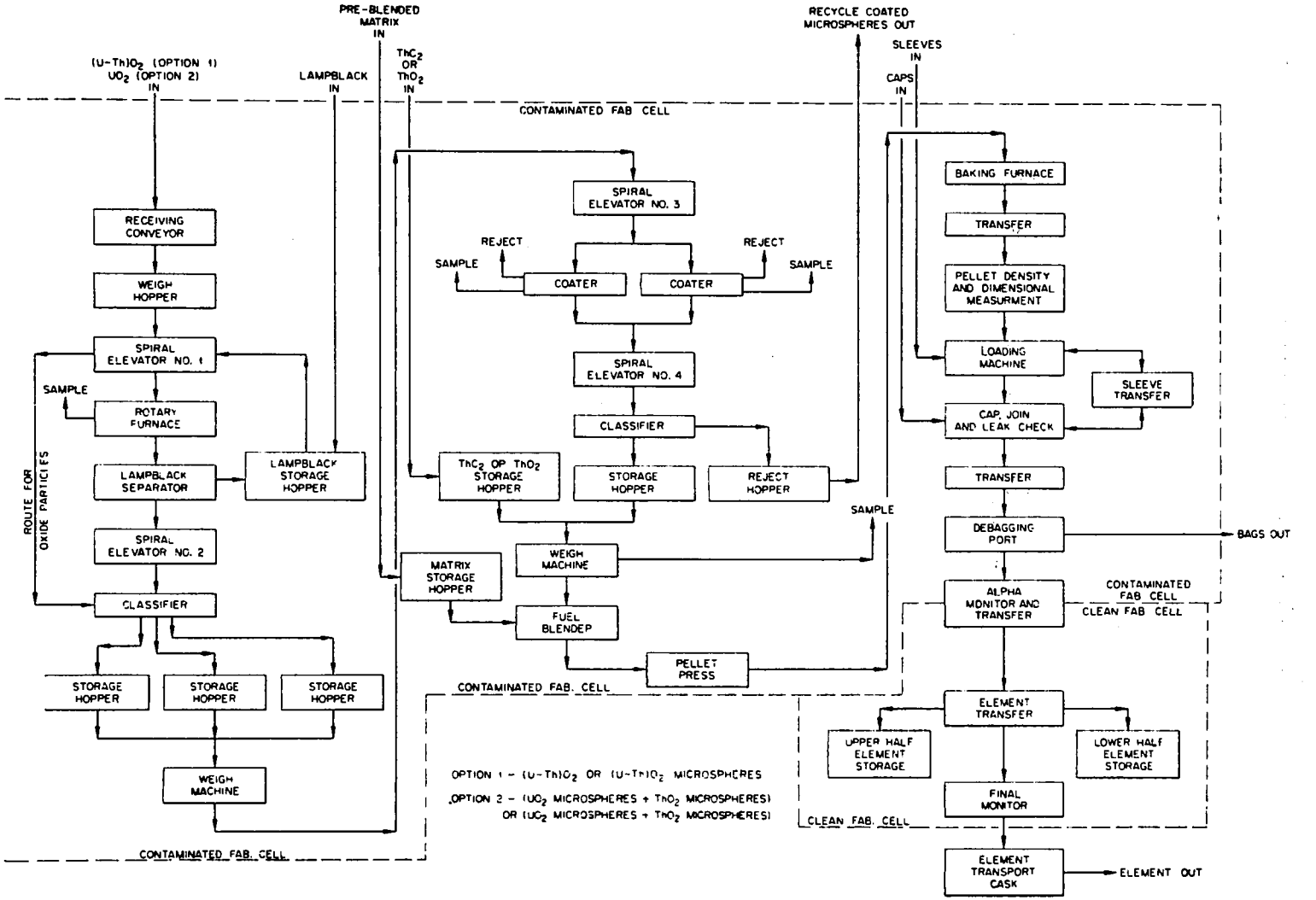


FIG. 14. — ORNL Thorium-Uranium Recycle Facility — Section Elevation.


PRE-BLENDED MATRIX IN
RECYCLE COATED MICROSPHERES OUT
SLEEVES IN
CAPS IN
(U-Th)O2 (OPTION 1) UO2 (OPTION 2) IN
LAMPBLACK IN
ThC2 OR ThO2 IN
CONTAMINATED FAB CELL
RECEIVING CONVEYOR
WEIGH HOPPER
SPIRAL ELEVATOR NO. 1
SAMPLE
ROTARY FURNACE
LAMPBLACK SEPARATOR
LAMPBLACK STORAGE HOPPER
ROUTE FOR OXIDE PARTICLES
SPIRAL ELEVATOR NO. 2
CLASSIFIER
STORAGE HOPPER
STORAGE HOPPER
STORAGE HOPPER
WEIGH MACHINE
CONTAMINATED FAB. CELL
SPIRAL ELEVATOR NO. 3
REJECT
SAMPLE
COATER
COATER
REJECT
SAMPLE
SPIRAL ELEVATOR NO. 4
CLASSIFIER
ThC2 OR ThO2 STORAGE HOPPER
STORAGE HOPPER
REJECT HOPPER
MATRIX STORAGE HOPPER
WEIGH MACHINE
FUEL BLENDER
PELLET PRESS
SAMPLE
BAKING FURNACE
TRANSFER
PELLET DENSITY AND DIMENSIONAL MEASURMENT
LOADING MACHINE
SLEEVE TRANSFER
CAP, JOIN AND LEAK CHECK
TRANSFER
DEBAGGING PORT
BAGS OUT
ALPHA MONITOR AND TRANSFER
CONTAMINATED FAB CELL
CLEAN FAB CELL
ELEMENT TRANSFER
UPPER HALF ELEMENT STORAGE
LOWER HALF ELEMENT STORAGE
FINAL MONITOR
CLEAN FAB. CELL
ELEMENT TRANSPORT CASK
ELEMENT OUT
OPTION 1 - (U-Th)O2 OR (U-Th)O2 MICROSPHERES
OPTION 2 - (UO2 MICROSPHERES + ThO2 MICROSPHERES) OR (UC2 MICROSPHERES + ThO2 MICROSPHERES)

manipulator system and the modular arrangement of windows and master-slave manipulators.

5.2. — REFABRICATION EQUIPMENT FOR HTGR FUEL.

We are now conceptually designing equipment to be used for demonstration of refabrication technology for the fueled graphite element depicted in Fig. 1. The flowsheet that is being used for the conceptual design is shown in Fig. 15 and is an expansion of the one presented in Fig. 3.

The equipment for demonstration of refabrication technology for fueled graphite elements will be of production type so that the information gained will be applicable both technically and economically to a full-scale production facility. The equipment is to be sized for the production of approximately 35 kg of heavy metal per day. The principal barrier to this goal, as far as capacity is concerned, is the coating process. We do not yet have a solution to the relatively low processing rate which is inherent to the coating step. Accordingly, we are exerting a substantial portion of our development program toward the solution of this problem.

A preliminary layout of the fabrication equipment in the TURF is shown in Fig. 16. The equipment will occupy approximately one-half of the space in the contaminated fabrication and clean fabrication cells. The dried sol-gel oxide microspheres are fed from the mechanical processing cell by means of a transfer conveyor (1) through the cell wall to the contaminated fabrication cell where they are charged to the inventory hopper (2). As required by the process, the particles are fed from the inventory hopper to the batch weighing device (3), then conveyed by the spiral elevator (4) to the rotary converter furnace (6). As the particles are charged to the spiral elevator (4), lamp black is dispensed from the lamp black storage hopper (5). After the reaction of the particles in the furnace, the lamp black and carbide particles are partitioned in the separator (7).

The carbide particles are then transferred to the top of the cell by means of the spiral elevator (8), and charged to the agglomerate separator (9), which separates particles that are stuck together. The material is stored in the storage hoppers (10). The material is dispensed from the storage hoppers by means of the batch weighing machine (11) and transferred vertically by the spiral elevator (12). The spiral elevator charges the coaters (13*a* and *b*) on a batch basis. The coated particles are removed from the coaters and fed to the spiral elevator (14) and through the elevator to the classifier (15) for separation of particle agglomerates. Material from the separator is fed to the storage hopper for useable material (16*a*); reject material is stored in another hopper (16*b*). [When particles bearing thorium alone are used, the material would be stored in a third storage hopper (16*c*)].

Batches of particles are dispensed by the weighing device (17) and mixed with the graphite flour, resin, binder, and solvent in the blender (19). The material which is mixed with the particles is preblended outside of the hot cell and charged to the storage hopper (18) by the cell manipulation system. This mixture is pressed in an automatic pellet press (20). After pressing, the green compacts are transferred to

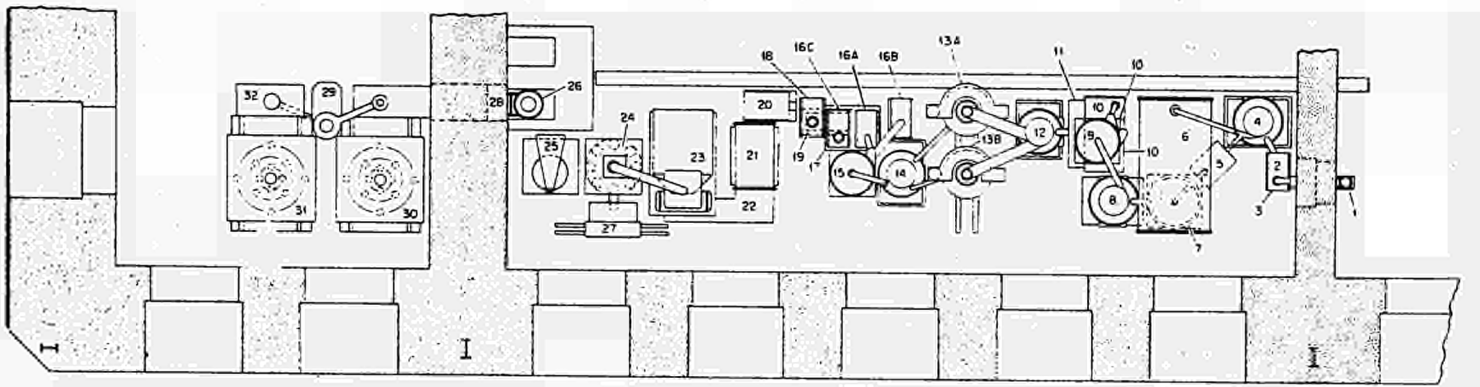


FIG. 16. — Layout of Refabrication Equipment in TURF.

the furnace (21) where they are baked at a temperature of 1,000° C. By means of the unloading conveyor (22), the pellets are discharged and removed to the inspection equipment (23). This inspection determines the pellet density and the conformity to the dimensional tolerances.

The pellets are loaded into the fuel channels in the graphite segments by the pellet loading machine (24). After loading, the segments are moved to the next machine (25) where they are capped with a graphite cap, sealed, and inspected. Following loading, the fuel segments are moved to the debagging port (26), where the protective covering for the elements is removed. Transfer operations involving the individual fuel segments are carried out by the fuel element transfer machine (27). After the fuel segment drops through the debagging port, it is placed in an alpha monitor (28) where it is monitored for contamination. Clean fuel segments are then moved into the clean fabrication cell by a transfer device (29). Fuel segments containing upper end cap fittings are placed in the upper storage area (30). Elements containing bottom end cap fittings are placed in the lower half element storage area (31). Before the fuel segment leaves the cell, it is monitored for gamma and alpha radiation (32).

Because of the handling limitations imposed by the building clearances, a fuel element longer than 12 ft cannot be handled in the facility; therefore, it will be necessary to accomplish the assembly operations for the fueled graphite element in another facility or at the reactor site. This could be done by means of a central fitting of the threaded or bayonet type, and the joint could be sealed with an inductively heated braze joint.

All of the hardware which is used in the fabrication of the fuel elements will be loaded into magazines and brought into the cell by means of the cell manipulation and crane system.

5.3. — Development Program.

To provide a basis for process and equipment design, we are conducting experiments in several areas including spherical particle preparation, conversion of oxide particles to carbide [23], and coating with pyrolytic carbon [24, 25, 26]. Most of this work is directed toward the scaleup of these processes. In fact, we are now equipping a pilotscale laboratory in which we shall accumulate data that we hope will lead to efficient remote fabrication processes and equipment. The facility is to process non-recycle material.

Summary

We have just begun to investigate the technology for refabrication of fueled graphite elements. Much more evaluation needs to be done and much thought must go into systems required for recycle of such fuel. However, it is encouraging that, despite the cost penalties associated with recycle of HTGR fuel, the concept shows promise for an economically attractive fuel cycle.

We have been reasonably optimistic in our cost analyses in that we have assumed that the sol-gel technique can be adapted satisfactorily to large production operations, that the conversion of sol-gel oxide microspheres to carbide on a large scale is within our grasp, and that some increase in the efficiency and capability of fluidized-bed coaters can be attained. It is clear that we must press development in these areas if we are to attain an economical thorium-uranium-233 recycle technology for high-temperature gas-cooled reactors.

We are conducting a comprehensive program at the Oak Ridge National Laboratory aimed at meeting these objectives. With the forthcoming pilot-scale demonstration of the sol-gel technique for making spherical particles, coupled with the coating process, we should have a better basis for the design of equipment for remote fabrication of the fuel in the TURF. Once equipment is operated in the TURF, we shall have a much better basis than we do now for the evaluation of the technical feasibility and cost of refabricating fueled graphite elements.

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SOME POSSIBILITIES OF AQUEOUS PROCESSING OF GRAPHITE-CARBIDE FUEL ELEMENTS

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ABSTRACT

In order to find a suitable head-end process permitting the treatment of graphite-carbide fuel elements in the aqueous processing cycle, a number of alternative methods for attacking graphite, pyrolytic carbon coatings and silicon carbide layers have been tested at Eurochemic. For practical reasons, the whole study has been restricted to fuels of two European reactors, namely the « Dragon » and the « Schulten » reactors.

These experiments have resulted in the presentation of two conceptual head-end flowsheets consisting essentially of :

— for the « Schulten » type fuel, the combustion of the carbon followed by fluoride catalyzed nitric acid leaching of the ashes, and,
— for the « Dragon » type fuel, the combustion of the carbon, followed by the combustion of the silicon carbide under the eutectic of the carbonates of lithium, sodium and potassium at 600° C, and by aqueous leaching, similar to the one described for the " Schulten " type fuel.

1. — INTRODUCTION

The general progress in development work on high temperature gas-cooled reactors emphasizes the lack of satisfactory reprocessing methods for graphite-based fuel elements. Currently considered are : (i) methods for mechanical, electrolytic or chemical disintegration of the graphite followed by or combined with nitric acid leaching : (ii) combustion of graphite and either acid leaching of the ashes or volatilization of the uranium by direct halogenation. While some of these possible processes show inherent drawbacks such as non-leachability of several percent of the uranium or the necessity of treating large volumes of gases, none has reached a development stage ripe for practical application. Thus the economic evaluation of fuel cycles for high temperature gas-cooled reactors using graphite fuel elements also remains incomplete. In particular, the controversy over fabrication of fission product releasing or fission product retaining fuel elements still continues. Thus continuous processing of a gas stream versus batchwise processing of the entire fuel elements is still in question.

Typical graphite fuel elements envisaged are those, for example, in the "Dragon" Reactor (1) and in the Schulten Reactor (2).

(1) High temperature Reactor Project Dragon, a joint undertaking of the OECD Nuclear Energy Agency.

(2) Experimental "potato-heap" reactor under construction at Jülich (Germany).

The former consist of cylindrical compacts of graphite covered by amorphous carbon ($\sim$ 45 mm diameter and $\sim$ 50 mm length) in which there are included either 300 μ diameter spherical particles with a uranium dicarbide-thorium dicarbide core covered with a silicon dicarbide layer followed by a pyrolytic carbon layer or, in a batch from a different zone of the reactor core, 300 μ particles with a uranium monocarbide-zirconium monocarbide core also coated by a silicon dicarbide layer and a pyrolytic carbon layer. The atomic ratio of the first batch U/Th/Si/C^{2-}/C is expected to be about 1/20/90/220/600, and for the second batch U/Zr/Si/C^{4-}/C about 1/8/90/9/180/600.

The elements of the Schülten type are graphite spheres, 60 mm diameter, containing 500 μ diameter spherical particles with a uranium dicarbide-thorium dicarbide core coated with pyrolytic carbon. The atomic U/Th/C ratio can be either 1/20/4,000 or 1/5/4,000.

In order to find a suitable head-end process permitting the *treatment of graphite-carbide fuel elements in the aqueous processing cycle*, a number of alternative methods for attacking graphite, pyrolytic carbon coating and silicon carbide layers have been tested at Eurochemic.

The experimental work reviewed here has been limited to some methods suitable for application to both types of European carbide-graphite fuels (Dragon and Schulten reactors).

2. — Selection of Systems for Attacking the Graphite, the Coatings and the Core on the Basis of the Known Bibliographic Data

2.1. — The Graphite and the Pyrolytic Carbon Coatings.

Two very distinct cases have to be considered :

— The graphite and the coated particles cannot be mechanically separated,

— The fuel is received as separated particles externally coated with pyrolytic carbon.

For the first type of elements, combustion with air or, if suitable, with nitrogen oxides, has been *selected* as the means of attacking the compacts instead of such methods as :

— grinding, [1], or,

— digestion with nitric acid with or without anodic disintegration [1].

Shortcomings of the combustion process have been noted by Ferris [1] such as :

— the limitation of the choice of materials for burner construction because of the high temperature involved;

— the preferential formation at high temperatures of CO, which could form explosive mixtures with oxygen, instead of CO_2, unless an excess of oxygen is present;

— the need for development of methods for safe disposal of radioactive off-gas;

— the need for using corrosive solutions (HNO_3 — HF) as leaching agent for the residues from thorium bearing particles;

— the difficulty of using a single vessel in the combustion and leaching steps, due to the markedly different thermal and environmental conditions in those two steps.

An attempt to simplify some of these problems by lowering the ignition temperature of the graphite with the aid of catalysts (such as manganese, copper or lead) has been described also by Ferris [1]. He reported that a treatment with 0.07 M lead acetate solution lowered the ignition point of graphite from 720° C to 345° C.

It was decided to *restrict* in the first stage the *experimental work* at Eurochemic to a comparison of the "dry" combustion of graphite with oxygen and with nitrogen dioxide with the same combustion in alkaline melts, which were chosen because they were already being tested as reagents for attacking silicon dicarbide (See 2.2).

For the second type of elements,

— *crushing,*

— *digestion with nitric acid, or,*

— *combustion*

are considered as means of attack, but only the last two have been selected for experimental testing at Eurochemic because crushing must be studied almost on a production scale to give significant results.

2.2. — The Silicon Carbide.

The silicon carbide coatings of the "Dragon" type fuel particles are said to consist of silicon dicarbide, while most of the published experimental work either deals especially with the monocarbide, i.e. : carborundum, or only refers to silicon carbide.

The pre-selection of systems to be tested for attacking silicon dicarbide was consequently based on bibliographic data on carborundum. This fact made it necessary to test systems that, according to the data on carborundum, had doubtful chances of success.

2.2.1. — *Reactions with gases.*

1. — Discussion.

(*a*) *Oxygen.*

It is expected that silicon dicarbide can be decomposed by oxygen, according to the reaction :

$$SiC_2 + 3O_2 \rightarrow SiO_2 + 2CO_2$$

which applies to complete combustion.

The oxygen for the combustion process could be supplied as such, as air or as a gaseous oxide giving oxygen through thermolysis.

J. A. Dillon, Jr. [2] carried out oxidation studies on silicon carbide in an atmosphere of oxygen and found that at temperatures above 900° C and pressures of about 1 atm. a silicon dioxide layer, which seems to be composed of hundreds of

monolayers, forms at the surface.

Jorgensen *et al* [3, 4] measured the rate of oxidation of silicon carbide in an atmosphere of dry oxygen between the temperatures of 900 and 1,600° C. The rate, determined with a thermobalance, was found to be diffusion controlled, the products of oxidation being amorphous silica or cristobalite depending on the temperature. The effect of reducing the impurity content of the silicon carbide from 1 % to 320 ppm. did not affect the rate of oxidation, but the vapour pressure and the partial pressure of oxygen were found to be extremely critical.

Lea [5] concludes from the study of more than twenty publications concerning the oxidation of silicon carbide that its oxidation begins in the neighbourhood of 800° C.

The following conclusions, cited by Lea and Jorgensen, and drawn by the authors of the references themselves are worth quoting :

— Muller and Baradue [6] reported that silicon carbide was not oxidized at 1,000° C and,

— Beecher and Hepburn [7] reported that the rate of oxidation had a maximum at 1,000° C.

— Elmer [8] who used an atmosphere of air in studying the rate of oxidation of silicon carbide powder by weight gain determinations, concluded that at 950 °C the rate was not diffusion controlled but was at higher temperatures, i.e. 1,400° C, that the products of oxidation were rapidly volatilized and that the rate controlling step was the growth of the SiO_2 layer by solid diffusion.

— Lambertson [9] in an unpublished report of the "Carborundum Company" agreed with Elmer in that the rate of oxidation was reaction controlled below 950° C; diffusion controlled between 950 and 1,650° C and reaction controlled above 1,650° C.

— Faust [10] stated that there are numerous reactions between silicon carbide and oxygen which are thermodynamically possible, even at room temperature, but in which the rate of oxidation does not become significant until temperatures in the neigbourhood of 900° C are reached.

(*b*) *Nitrogen oxides.*

The nitrogen oxides release oxygen by thermal decomposition. It is known, for instance :

— that nitrogen dioxide decomposes according to the reaction :

$$NO_2 \rightleftarrows NO + \frac{1}{2} O_2,$$

— that this reaction is 25 % displaced towards the decomposition products at 300° C and 75 % at 500° C [11], and,

— that the thermolysis of nitric oxide [12] takes place according to :

$$NO \rightleftarrows \frac{1}{2} N_2O + \frac{1}{2} O$$

$$N_2O \rightleftarrows N_2 + \frac{1}{2} O_2$$

It appears that, if the reaction of silicon carbide with oxygen is thermodynamically possible [10] its reaction with NO_2 must also be possible, but the temperature at which the rate of reaction becomes significant remains to be determined.

(*c*) *Halogens.*

It has been reported [13] that fluorine readily reacts with silicon carbide.

Work has also been reported on the reaction of chlorine with silicon carbide [14] :

— it reacts at temperatures as low as 600° C.
— up to 900° C, $SiCl_4$ is formed leaving a layer of carbon on the surface,
— above 900° C, both CCl_4 and $SiCl_4$ are formed leaving a surface free from products of reaction.

2. — Conclusions.

The following conclusions were drawn as guides to the experimental work on the removal of silicon dicarbide layers by gases.

(*a*) *Oxygen* (*or air*).

The effect of oxygen (or air) on silicon dicarbide layers had to be experimentally studied at temperatures from 700-1,200° C because :

— the silicon dicarbide coated particles ("Dragon" type fuel) are inseparable from the graphite matrix by mechanical means, and in any case coated with a pyrolytic carbon layer, and,
— the straight combustion of graphite would lead to temperatures of at least 800° C; actually Ferris [15] has reported 1,450° C as the probable temperature reached by the graphite.

(*b*) *Nitrogen oxides.*

The effect of nitrogen dioxide on silicon dicarbide coatings was considered to be worth studying experimentally from temperature as low as 300° C.

(*c*) *Halogens.*

These were considered attractive as a basis for processes attacking silicon dicarbide without leaving residual solids.

Their experimental study was left until later so that means could be sought of providing milder corrosion conditions than those which would result from direct use of elemental fluorine or chlorine while keeping the principle of the volatilization of a silicon halide.

2.2.2. — *Reactions with Molten Salts.*

1. — Discussion.

(*a*) *Alkaline hydroxides.*

Molten alkali hydroxides are [16] :

— strongly dissociated, according to : $MOH \rightarrow M^+ + OH^-$, and,
— strong O^{2-} donors according to : $2OH^- \rightarrow O^{2-} + H_2O$

It was expected that in the presence of oxygen (supplied as such, as air or as a compound unstable at the temperature of the bath and releasing oxygen by its decomposition) the silicon dicarbide coatings would be attacked in a bath of molten alkali hydroxides through a process consisting of :

— the oxidation by oxygen, leading to SiO_2 and CO_2;

— the neutralization of CO_2 and its dissolution in the bath as CO_3^{2-}, according to the reaction [16] :

$$CO_2 + O^{2-} \underset{\leftarrow}{\rightarrow} CO_3^{2-}$$

— the neutralization of the SiO_2 and its dissolution as SiO_3^{2-}, SiO_4^{4-}, $Si_2O_5^{2-}$, $Si_2O_7^{6-}$ and polysilicates of general formula $(SiO_3)_n^{2n-}$ [16] according to the reactions :

$$SiO_2 + O^{2-} \underset{\leftarrow}{\rightarrow} SiO_3^{2-}$$

$$SiO_2 + 2O^{2-} \underset{\leftarrow}{\rightarrow} SiO_4^{4-}$$

$$2SiO_2 + O^{2-} \underset{\leftarrow}{\rightarrow} Si_2O_5^{2-}$$

$$2SiO_2 + 3O^{2-} \underset{\leftarrow}{\rightarrow} Si_2O_7^{6-}$$

$$nSiO_2 + nO^{2-} \underset{\leftarrow}{\rightarrow} (SiO_3)_n^{2n-}$$

If the reaction of neutralization of SiO_2 is simplified to the formation of orthosilicate, SiO_4^{4-}, the expected overall process can be expressed by :

$$SiC_2 + 3O_2 + 8OH^- \rightarrow SiO_4^{4-} + 2CO_3^{2-} + 4H_2O.$$

If the reaction of neutralization of SiO_2 is simplified to the formation of metasilicate, SiO_3^{2-}, the expected overall process can be expressed by :

$$SiC_2 + 3O_2 + 6OH^- \rightarrow SiO_3^{2-} + 2CO_3^{2-} + 3H_2O.$$

With NaOH at 900° C an etching rate of silicon carbide of 0.5 mg/cm²/min has been reported [10]. The same author reported that after 45 min at 500° C, no etching of silicon carbide by NaOH was observed.

(*b*) *Alkaline carbonates.*

Molten alkali carbonates are [16] :

— dissociated according to :

$$M_2CO_3 \rightarrow 2M^+ + CO_3^{2-}, \qquad \text{and,}$$

— O^{2-} donors according to :

$$CO_3^{2-} \underset{\leftarrow}{\rightarrow} O^{2-} + CO_2$$

It was expected that the silicon dicarbide coatings would be attacked in a bath of molten alkali carbonates in the presence of oxygen by a process similar to the one described for the hydroxides, but in which the CO_2 would not be neutralized and retained in the melt.

The expected overall process can be expressed by :

$$SiC_2 + 3O_2 + 2CO_3^{2-} \rightarrow SiO_4^{4-} + 4CO_2$$

or by :

$$SiC_2 + 3O_2 + CO_3^{2-} \rightarrow SiO_3^{2-} + 3CO_2$$

according to whether the formation of ortho-or metasilicates is assumed.

At 900° C with Na_2CO_3(m.p. 850 °C) an etching rate of silicon carbide of 0.1 mg/cm²/min has been reported [10].

2. — Conclusions.

Both types of attack appeared feasible.

(*a*) *Alkaline hydroxides.*

The attack in molten alkaline hydroxides had the short-coming for being applied as a decoating process of giving a high theoretical ratio M^+/Si [6-8] by the neutralization of the CO_2; but the introduction of OH^- into a carbonate bath for the start up period might be of interest and some experimental tests with this type of bath seemed appropriate.

(*b*) *Alkaline carbonates.*

A process for attacking silicon dicarbide in molten alkaline carbonates appeared more attractive than in alkaline hydroxides, as the ratio M^+/Si produced by the melt could be theoretically as *low* as 4 or 2, leading to a lower M^+/UO_2^{2+} ratio in the solution prepared by subsequent aqueous leaching.

The eutectic of the carbonates of lithium, sodium and potassium (m.p. 397° C) was selected as bath for the experimental work. The real M^+/Si ratio in either type of decoating process will be determined :

— by the maximum desirable temperature for the process, and,

— by the solubility of Si in the bath (carbonates or carbonates-hydroxides mixture) at that temperature.

2.2.3. — *Reactions with solutions.*

1. — Discussion :

The attack of silicon carbide requires the formation of compounds of both components, silicon and carbon.

Consequently, silicon carbide is strongly resistant to chemical attack below temperatures of several hundreds of degrees : the difficulty of forming compounds from any of the forms of carbon is well known.

J. W. Faust [10] reviewed the possibilities of etching carborundum with aqueous solutions of acids.

The conclusion on the possibilities of etching with HNO_3-HF was that, although thermodynamic calculations show that HNO_3 solution can oxidize carbon at room temperature, the activation energy is too great for the reaction to take

place even at 90° C, as shown by the fact that SiC neither gains nor loses weight after being in a solution of HF and HNO_3, at 90° C for 4 hours.

Wunder and Jeanneret [17] reported that SiC heated in phosphoric acid at 230° C was completely decomposed in about 3 hours the resulting solution containing a white gelatinous precipitate, which seemed to be silica, while the carbon was in suspension.

2. — Conclusion.

While it appeared that none of the possible etching reagent for silicon is potent enough to form compounds with carbon, it was decided to screen rapidly the attack with aqueous solutions of several mineral acids, to test the possibilities of mechanical disintegration of the coatings due to the etching of the silicon.

2.3. — The Carbides of the Core.

The type of aqueous leaching of the core of the coated particles depends not only upon the nature of the core itself (uranium-thorium dicarbide or uranium-zirconium monocarbide), but also on the system used for the attack of :

— the graphite matrix and/or the pyrolytic carbon coating, and,
— the silicon dicarbide layer, if present ("Dragon").

Crushing methods lead to the leaching problem of dissolving :

— either, uranium-thorium dicarbide,
— or, uranium-zirconium monocarbide.

"*Dry*" *Combustion methods* lead to the dissolution of :

— either U_3O_8-ThO_2, accompanied by SiO_2 in the case of silicon dicarbide coatings submitted to temperatures high enough to bring about its combustion.
— or, U_3O_8-ZrO_2, with SiO_2 as well in the same conditions as above.

Some physical properties of these oxides, which have an influence on the reactivity toward aqueous reagents, could be affected by the temperature reached during the combustion of the carbide form. Interest therefore exists in keeping that temperature as low as possible.

Combustion of the silicon carbide layer inside baths of molten carbonates is expected to lead to the dissolution of :

— either a mixture of thoria and alkaline uranates, silicates and carbonates.
— or, a mixture of zirconium oxide (or possibly even zirconate), with alkaline uranates, silicates and carbonates.

It was decided to postpone any experimental work on the possibilities of aqueous leaching of the core of the particles until a defined system for the decoating has been selected which will fix the nature of the ashes.

A review of the characteristics of the systems of aqueous leaching, according to the nature of the ashes is given in the following account.

2.3.1. — *Dissolution of the Untransformed Decoated Carbide Core.*

The dissolution of the carbides of uranium, thorium and zirconium does not represent a major problem, according to the extensive work carried out at different laboratories, mainly at Oak Ridge.

(*a*) Uranium Carbides.

It has been reported that *uranium dicarbide* is hydrolysed by water [18, 19] its reaction at boiling point or even at 80° C being immediate and leading to a mixture of about 23 % H_2, 14 % CH_4, 38 % C_2H_6 and 22 % hydrocarbons containing 3 to 8 atoms of carbon [19, 20, 21].

Results have also been reported on the hydrolysis of *uranium monocarbide* by water at 80° C [21] giving 10.4-12.5 % H_2, 86.7-81.1 % CH_4, 1.8-5.4 % C_2H_6, 1.1 % C_3-C_8 hydrocarbons and on its dissolution with 4-5 M HNO_3 [20-23] producing a strongly coloured solution from which UO_2^{2+} was sorbed on a cationic resin and the strongly coloured species on an anionic resin. The off-gas near the end of the second reaction was 65 % HNO_3, 23 % CO_2, 4 % nitrous oxide, 1 % CO and 5 % N_2 (atmospheric contaminant)- Reports must also be noted :

— that hydrolysis of UC by water at 80° C yielded a finely divided, insoluble uranium compound and 92 cc of gas per gram of UC [20-23] and,

— that hydrolysis of UC specimens containing 20 % UC_2 yielded some wax.

(*b*) Thorium Carbide.

The hydrolysis of *thorium dicarbide* by water was reported as a complex reaction, one of the features of which was the production of substantial amounts of liquid and solid hydrocarbons in addition to the formation of a hydrated oxide [16]. However, it has more recently been reported that thorium dicarbide on hydrolysis yielded methane almost exclusively [24].

(*c*) Zirconium Carbide.

Hot concentrated nitric acid reacts violently [25] with *zirconium monocarbide* following, according to Blumenthal [26], an oxidation reaction.

Diluted or concentrated hydrofluoric acid attacks the carbide [27,28] following, according to Blumenthal [26] a neutralisation reaction, the liberated atoms of carbon reacting partially with the nascent hydrogen to form hydrocarbons.

2.3.2. — *Dissolution of the Oxides Formed by "Dry" Combustion.*

The dissolution of the oxides of uranium, thorium and zirconium depends very much on the temperature at which they have been formed.

(*a*) Uranium Oxide.

The dissolution of the U_3O_8 with HNO_3 is not a problem by itself, but the fraction of uranium recovered from the mixed U_3O_8-ThO_2, U_3O_8-ThO_2-SiO_2, U_3O_8-ZrO_2, or U_3O_8-ZrO_2-SiO_2 oxides depends on the particular conditions of formation.

(*b*) Thorium Oxide.

Calcined at high temperature, it is soluble in concentrated nitric and hydrofluoric acids separately [20]. Burgers and Van Liempt [29] observed only a slight attack by a HNO_3-HF mixture.

On the other hand, Schuler, Steahly and Stoughton [30] reported a faster dissolution in concentrated HNO_3 containing small amounts of fluoride or fluosilicate, whose presence catalyzes the dissolution, probably by the formation of thorium fluoride complexes. The oxide formed at low temperature is more easily soluble, due, according to Foëx [31] to the size of the cristallites. Experimental work at Oak Ridge on the dissolution of ThO_2 and ThO_2-UO_2 has shown that, all the other variables being constant, the rate of dissolution is a maximum with 13 M HNO_3-0.06 M HF [32, 33, 34]. It is still not known what fraction of thorium is retained in the silica, when it is present in large amounts, i.e. during the reprocessing of "Dragon" fuel.

(*c*) Zirconium Oxide.

The reagent used for dissolution of uranium and thorium will leave the zirconium as insoluble oxide. Uranium and thorium oxides form solid solutions with zirconium oxides and, as a consequence, it is probable that they will be retained in the insoluble zirconium oxide during the leaching with mineral acids.

2.3.3. — *Dissolution of the Residue from Combustion of the Silicon Carbide Layer inside Baths of Molten Carbonates.*

It is expected that suitable reagents will be similar to those serving for the dissolution of the ashes formed in "dry" combustion, i.e. 13 M HNO_3 — 0.06 M HF.

The process, which consists of the neutralization of the carbonates, silicates, uranates and thorium oxide with evolution of CO_2, formation of a solution of alkaline, uranyl and thorium nitrates and precipitation of silica and zirconium oxide *is expected to improve the uranium and thorium recovery* from either the silica or the mixed silica-zirconium oxide, due to :

— the relatively low temperature of combustion (600° C) compared with the temperature of the "dry" combustion, and,
— the complete transformation of the melt during the leaching by the aqueous acid solution.

3. — Experimental results

3.1. — The silicon Carbide and the Pyrolytic Carbon Coatings.

The combination of the experimental study on the removal of the coats of pyrolytic carbon and silicon dicarbide appeared convenient from a practical standpoint : the "mock up" silicon dicarbide coated particles available for the study consisted

of a uranium-thorium dicarbide core coated by layers of pyrolytic carbon, silicon dicarbide and again pyrolytic carbon.

The main effort has been concentrated upon the combustion with oxygen (or air) and with nitrogen dioxide,

— either in a "*dry*" *combustion* process,
— *or*, in a combustion *in a molten bath*, after
— a *wide screening* of chemical reagents which will be described later as "other experiments " and which have given negative results,
— *an unsuccessful attempt* at "mechanical " cracking of the particles by methods not requiring direct contact of the particles with mechanical tools, i.e. induction or dielectric heating.

3.1.1. — *Experimental Techniques.*

(*a*) Thermolysis in "dry" atmospheres of oxygen or nitrogen dioxide.

The oxidation in atmospheres of oxygen, air or nitrogen dioxide of "mock-up" "Dragon" type fuel core particles ([1]) has been studied using a fused quartz spring thermobalance (Fig. 3.1.1.-1-).

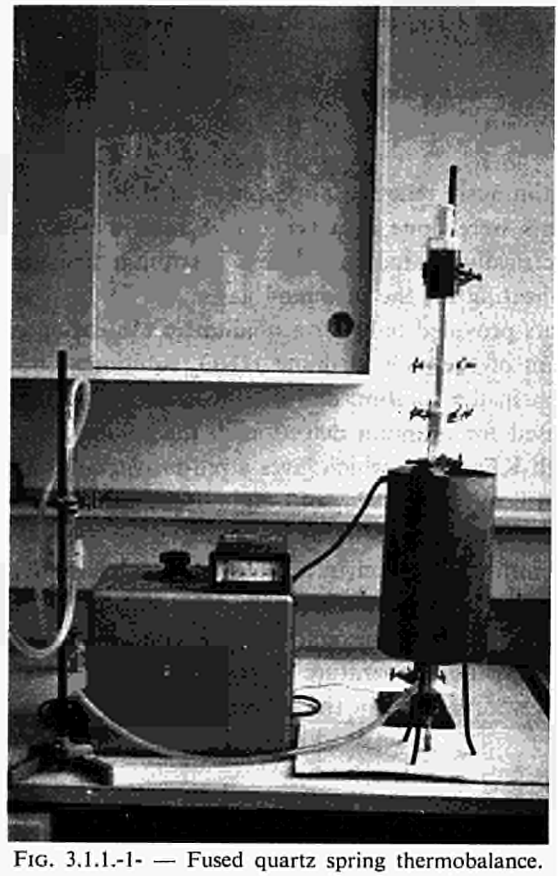

FIG. 3.1.1.-1- — Fused quartz spring thermobalance.

The maximum temperature attainable in the combustion tube of the furnace, a vertical 28 mm internal diameter quartz cylinder, was 1,100° C, measured with a prevision of ± 10° C. The sensitivity of the balance, consisting of a quartz spiral whose extension was measured by a cathetometer, ranged from 0.03 to 0.06 mg, depending on the spring used.

The experiments consisted of determining the weight of sets of particles as a function of the temperature in streams of oxygen (or air) or nitrogen dioxide at controlled flow rates.

The different types of particles used for the experiments are described in Table 3.1.1.-1-.

(*b*) Thermolysis in molten alkaline baths.

A study was made of the treatment of fuel particles in molten alkaline baths in atmosphere of oxygen (or air) and nitrogen dioxide. The same kind of "mock-up" "Dragon" type fuel particles were used as in the preceding experiments.

Experiments were performed with the eutectic mixture of the carbonates of lithium, sodium and potassium (43.5 % Li_2CO_3, 31.5 % Na_2CO_3, 25 % K_2CO_3) as molten bath (m.p. 397° C) at temperatures from 400 to 900° C, and also with molten caustic soda.

The particles immersed in the bath were submitted to the different temperatures :
— either with continuous sparging of the melt with oxygen (or air) or nitrogen dioxide,
— or under a continuously renewed atmosphere of those gases.

The experiments were done in a vertical 78 mm internal diameter cylindrical nickel reactor, electrically heated to reach a maximum temperature of 1,100° C, provided with pre-heating for the incoming gases.

The experiments provided only for a qualitative evaluation of the effects of the treatment-dissolution of the melt in dilute HNO_3 solution, followed by qualitative analyses of uranium in the solution and of silicon in the solid residue.

The method used for uranium detection in the solution consisted of a colorimetric spot test with $K_4Fe(CN)_3$, which gives a red-brown coloration or precipitation with a limit of identification of 0.92 γ of uranium in a limit of dilution $1/5 \times 10^4$.

The detection of silicon in the residue was based on the following tests :
— washing of the residue, ignition in a platinum crucible and weighing,
— treatment of the ignited residue with 30 % HF solution and weighing after ignition, the weight difference corresponding to silicon.

The limitations on the temperature of the process due to the solubility of the silicates in the bath were studied in the following way.

---

(1) Obtained from the Metallurgical Laboratories of the High Temperature Reactor Project "Dragon".

TABLE 3.1.1. 1. — Characteristics of the "mock-up" spherical particles used in the experiments

Characteristics	Type			
	A	B	C	D
External diameter	300 μ	Same as A	Same as A	Same as A
Core	UC_2	Same as A	ThC_2-UC_2(Th/U = 15)	ThC_2-UC_2(Th/U = 4)
Coatings : from the core to the external surface.	60 μ pyrolytic C	3 μ pyrolytic C 45 μ SiC_2 40 μ pyrolytic C	15 μ pyrolytic C 30 μ SiC_2 50-55 μ pyrolytic C	Same as C Same as C Same as C
Compositon :				
Uranium carbide	Not given	24 w. %	Not given	Not given
Thorium carbide	Not given	—	Not given	Not given
Silicon carbide	Not given	28 w. %	Not given	Not given
External pyrolytic carbon coating	Not given	48 w. %	Not given	Not given

A eutectic mixture of carbonates of lithium, sodium and potassium was mixed with :
— either a mixture of metasilicates of lithium, sodium and potassium in which the Li/Na/K ratio was the same as in the eutectic of the carbonates,
— or, a mixture of orthosilicates of lithium, sodium and potassium of the same Li/Na/K ratio,
— or silica,
— or carborundum.

In this way different Si/CO_3^{2-} ratios were obtained.

Before the solubility curves were determined, the first three mixtures were held for long periods at temperatures at which the whole mixture was in the liquid state, and the fourth mixture was melted and continuously sparged with oxygen in order to obtain the thermolysis of the carborundum inside the melt.

In all the cases the treatment was carried out in the nickel reactor already described.

A first estimation of the solubility was made by submitting 10 g aliquots of the mixtures after the treatment to a temperature lower than that at which total dissolution was expected, the samples being contained in nickel crucibles. After 1 hour the samples were visually examined and the temperature was raised by 20° C steps until complete dissolution was observed. The temperature was then decreased by the same short steps until precipitation started to be observed. The temperature was controlled in the combustion chamber near the crucibles with a precision of $\pm$ 20° C, but not in the melt itself.

The results reported were determined in this way.

The same method was used to determine the freezing curves of ternary mixtures of alkali hydroxides-carbonates and the silicates obtained by combustion of carborundum under the eutectic mixture of lithium, sodium and potassium carbonates.

There was a variable content of OH^-, constant initial Si/CO_3^{2-} ratio of 0.3 and overall $Li^+/Na^+/K^+$ ratio equal to that of the eutectic of the carbonates.

The method was considered acceptable for the purpose of a first estimation of the limitations of such processes; for accurate determination of the freezing curves, precautions have to be taken such as :
— the containment of the bath in a covered nickel crucible provided with a heating jacket filled with a salt of a melting point lower than the temperatures of the bath, the eutectic of the carbonates for instance,
— the immersion in the bath itself of the nickel/nickel chrome thermoelement protected by a "Degussit" canning.

(*c*) Effects of induction or dielectric heating.

The same type of "mock-up" "Dragon" fuel particles used in the previous experiments were submitted to a very rapid rise of temperature by induction or dielectric heating using high frequency current generators.

By courtesy of "Himmelwork" a set of experiments was carried out in their High Frequency Generators Department (Tübingen, Germany). The purpose of

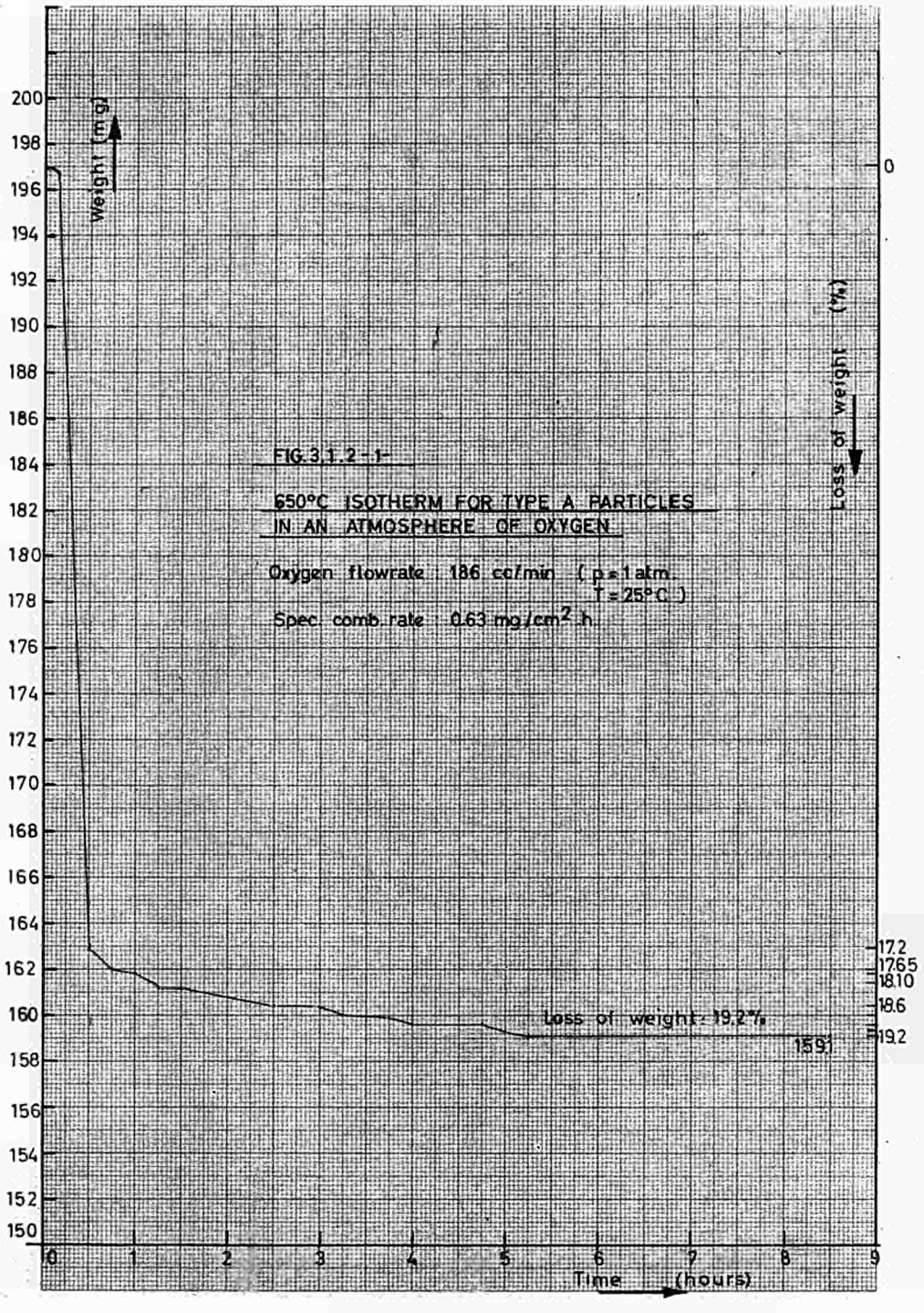
Weight (mg)
200
198
196
194
192
190
188
186
184
182
180
178
176
174
172
170
168
166
164
162
160
158
156
154
152
150
0
1
2
3
4
5
6
7
8
9
Time (hours)
Loss of weight (%)
0
17.2
17.65
18.10
18.6
19.2
Loss of weight: 19.2%
159
FIG. 3.1.2.-1-
650°C ISOTHERM FOR TYPE A PARTICLES IN AN ATMOSPHERE OF OXYGEN
Oxygen flowrate: 186 cc/min (p = 1 atm. T = 25°C)
Spec. comb. rate: 0.63 mg/cm² h

FIG. 3.1.2.-1-

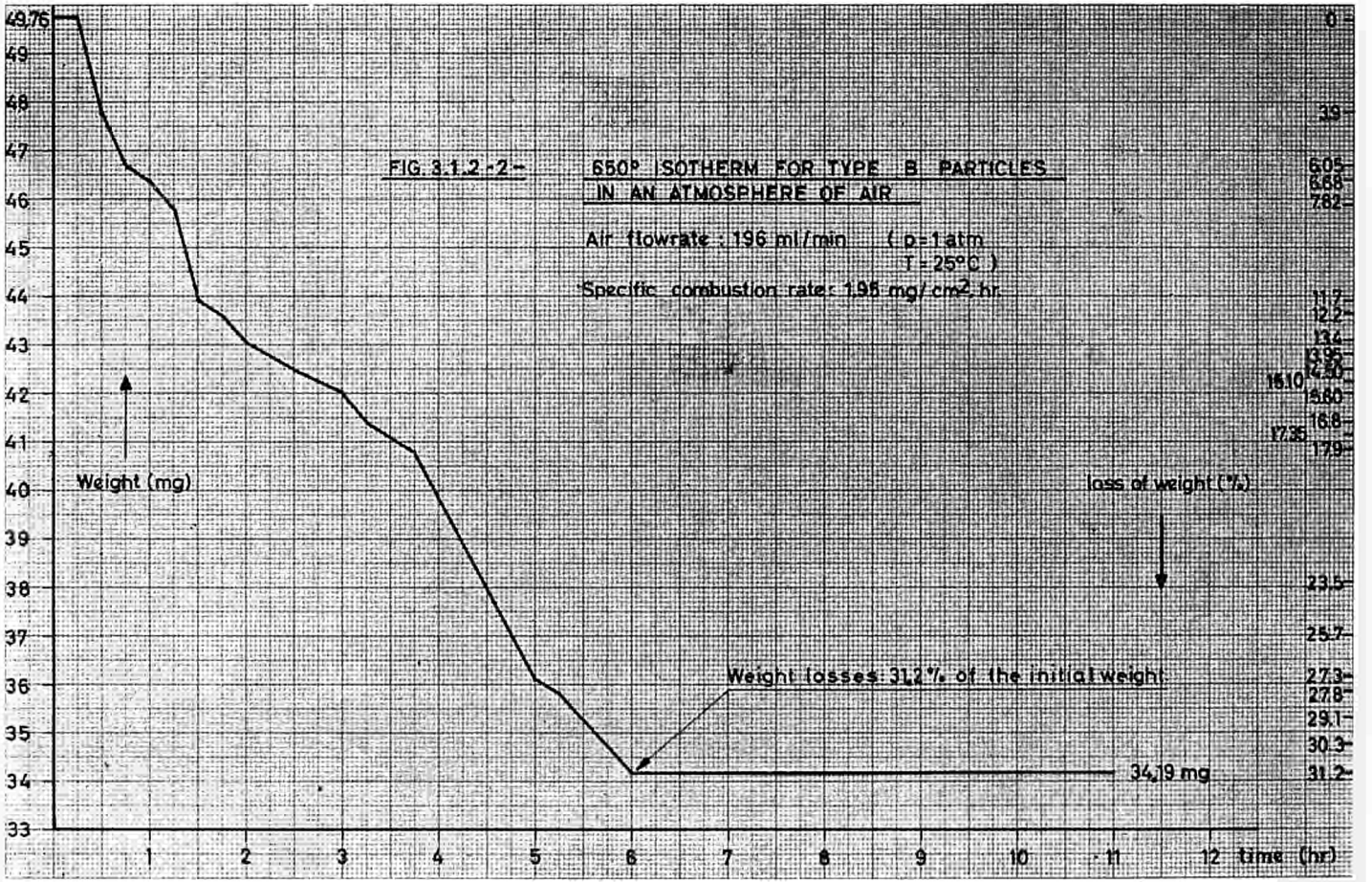
FIG. 3.1.2.-2-
650° ISOTHERM FOR TYPE B PARTICLES IN AN ATMOSPHERE OF AIR
Air flowrate : 196 ml/min (p=1 atm T = 25°C)
Specific combustion rate: 1.95 mg/cm2.hr
Weight (mg)
loss of weight (%)
Weight losses: 31.2 % of the initial weight
34.19 mg
time (hr)
49.76
0
39
6.05
7.62
11.7
12.2
13.4
13.95
14.50
15.10
15.60
16.8
17.35
17.9
23.5
25.7
27.3
27.8
29.1
30.3
31.2

FIG. 3.1.2.-2-

these qualitative experiments was to observe whether thermolysis or mechanical cracking of the silicon carbide layer was possible by these methods, leading to the development of a "crack and leach" process.

The experiments were carried out with :

— an inductive heat generator with 2 kW output power and 2 Megacycles/sec frequency,
— a dielectric heat generator with 1 kW output power and 27 Megacycles/sec frequency,
— a dielectric heat generator with 2 kW and 30 Megacycles/sec.

After the high frequency treatment, the particles were sectioned and micrographed by the Metallurgical Department Service of the C.E.N. (Mol, Belgium).

(*d*) Other experiments.

The same type of "mock-up" "Dragon" type fuel core particles used in the experiments described in all the preceding sections were submitted to the action of the following reagents :

— boiling nitric acid solutions of concentrations from dilute to fuming,
— boiling concentrated sulfuric acid or persulfuric acid in the presence of silver ions,
— boiling concentrated chromic acid,
— boiling concentrated perchloric acid,
— boiling phosphoric acid,
— boiling concentrated hydrochloric acid,
— boiling concentrated hydrofluoric acid.

After prolonged treatment and cooling, the particles were simply examined visually and in the cases of obvious disintegration the qualitative tests described in 3.1.1. (*b*) (on uranium in the solution and silicon in the residue) were made.

3.1.2. — *Results.*

(*a*) Thermolysis in "dry" atmosphere of oxygen or nitrogen dioxide.

Figs 3.1.2.-1, 2, 3, 4- give gravimetric isotherms for "dry" combustion in oxygen for the particles of the types A, C and D and in air for the type B.

The weight loss of the type A at 650° C (Fig. 3.1.2.-1-) can be explained by a complete combustion of the pyrolitic carbon layer and the uranium dicarbide core leading to a final U_3O_8 form : the residue dissolved in boiling 10 M HNO_3 and the resulting solution gave positive tests for uranium.

The weight losses of the other three types at 650° C (Fig. 3.1.2.-2-), 1,000° C (Fig. 3.1.2.-3-), and 1,050° C (Fig. 3.1.2.-4-) are in agreement with a combustion of the external pyrolytic carbon coating, and this is supported by the undestroyed appearance of the particles and negative results of U or Si detection tests in the solution resulting from the leaching of the particles by 30 % HF solution.

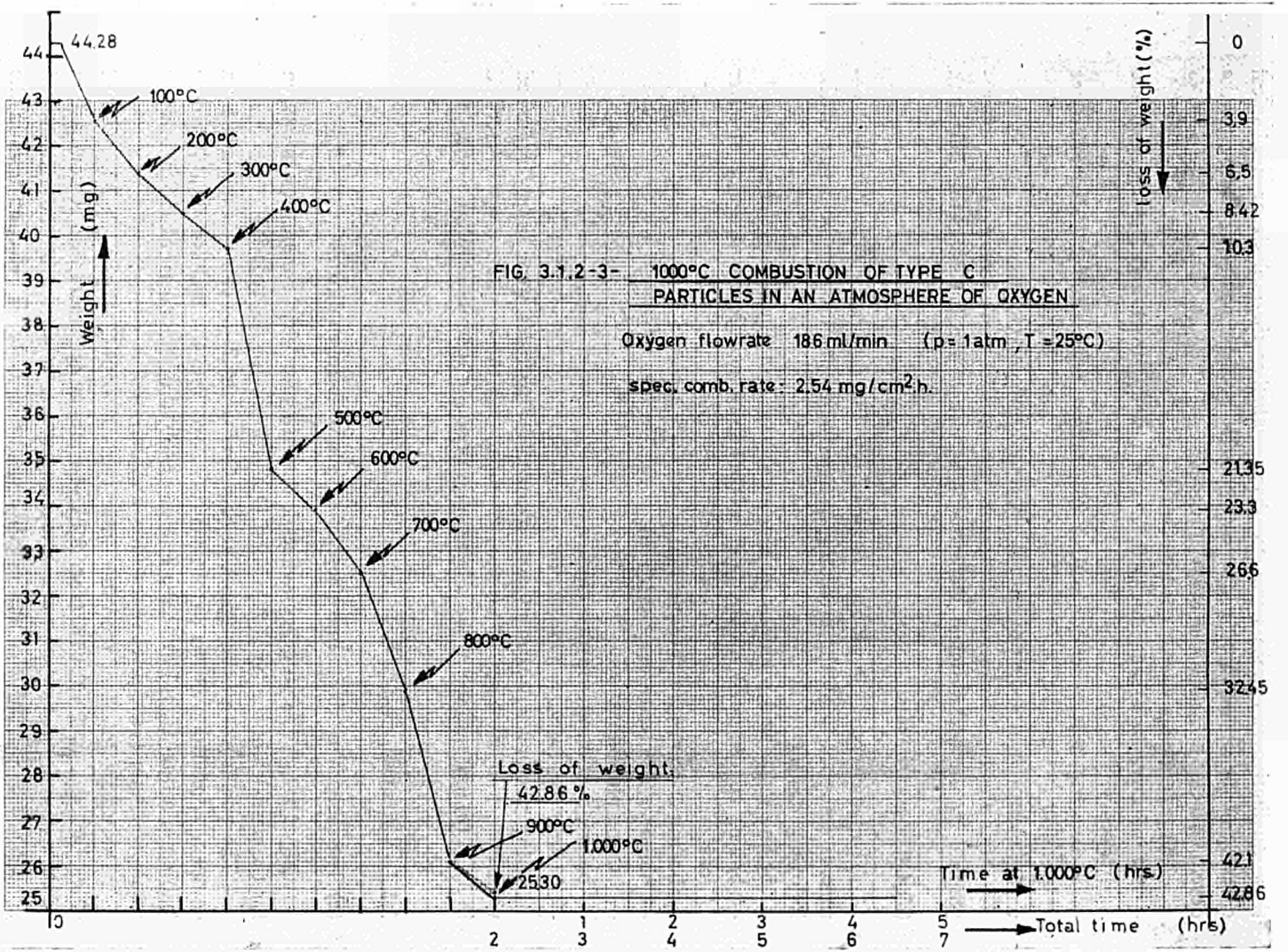
44.28
100°C
200°C
300°C
400°C
500°C
600°C
700°C
800°C
900°C
1.000°C
25.30
Loss of weight
42.86 %
Weight (mg)
Loss of weight (%)
0
3.9
6.5
8.42
10.3
21.35
23.3
26.6
32.45
42.1
42.86
Time at 1.000°C (hrs.)
Total time (hrs)
FIG. 3.1.2-3- 1000°C COMBUSTION OF TYPE C PARTICLES IN AN ATMOSPHERE OF OXYGEN
Oxygen flowrate 186 ml/min (p = 1 atm, T = 25°C)
spec. comb. rate: 2.54 mg/cm².h.

FIG. 3.1.2.-3-

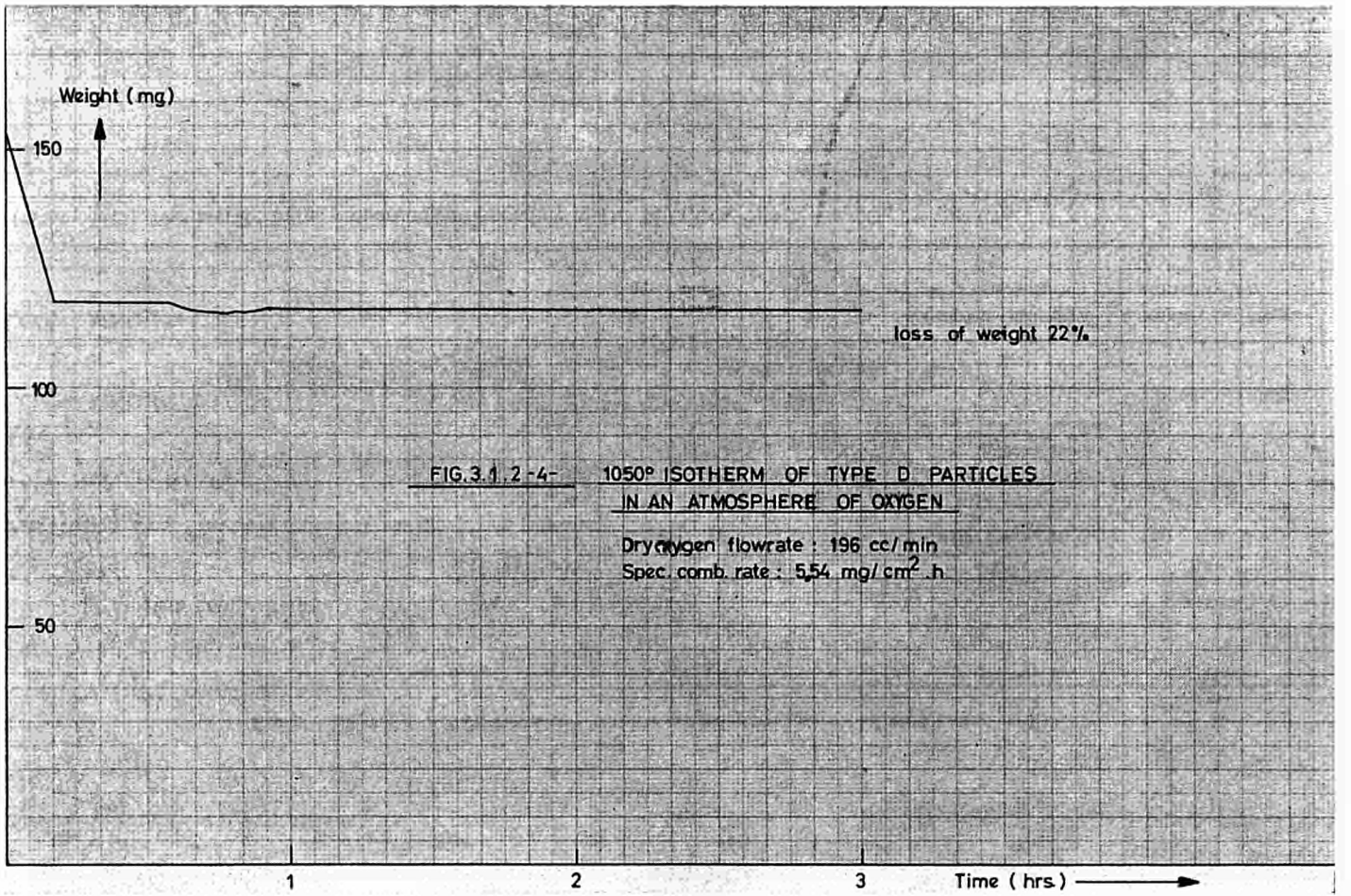
Weight (mg)
150
100
50
loss of weight 22%
FIG. 3.1.2-4- 1050° ISOTHERM OF TYPE D PARTICLES IN AN ATMOSPHERE OF OXYGEN
Dry oxygen flowrate : 196 cc/min
Spec. comb. rate : 5,54 mg/cm^2.h
1
2
3
Time (hrs.)

FIG. 3.1.2.-4-

TABLE 3.1.2.-1. — Attack of silicon carbide coated particles in molten alkaline baths

Bath composition	Results at different temperatures			
	400° C	500° C	600° C	700° C
50 % w. NaOH — $NaNO_3$	No attack	Complete attack after 2 h	Complete disintegration after 1 h	
M_2CO_3 — MNO_3 (*), $CO_3^{2-}/NO_3^- = 4$		Not complete disintegration after 5 h		Complete disintegration after 1 h
M_2CO_3 — MNO_3 (*), $CO_3^{2-}/NO_3^- = 1$			Complete disintegration after 4 h	Complete disintegration after 2 h

(*) $M^+ = Li^+$, Na^+ and K^+ in the proportion corresponding to 43.5 % Li_2CO_3, 31.5 % Na_2CO_3, 25 % K_2CO_3.

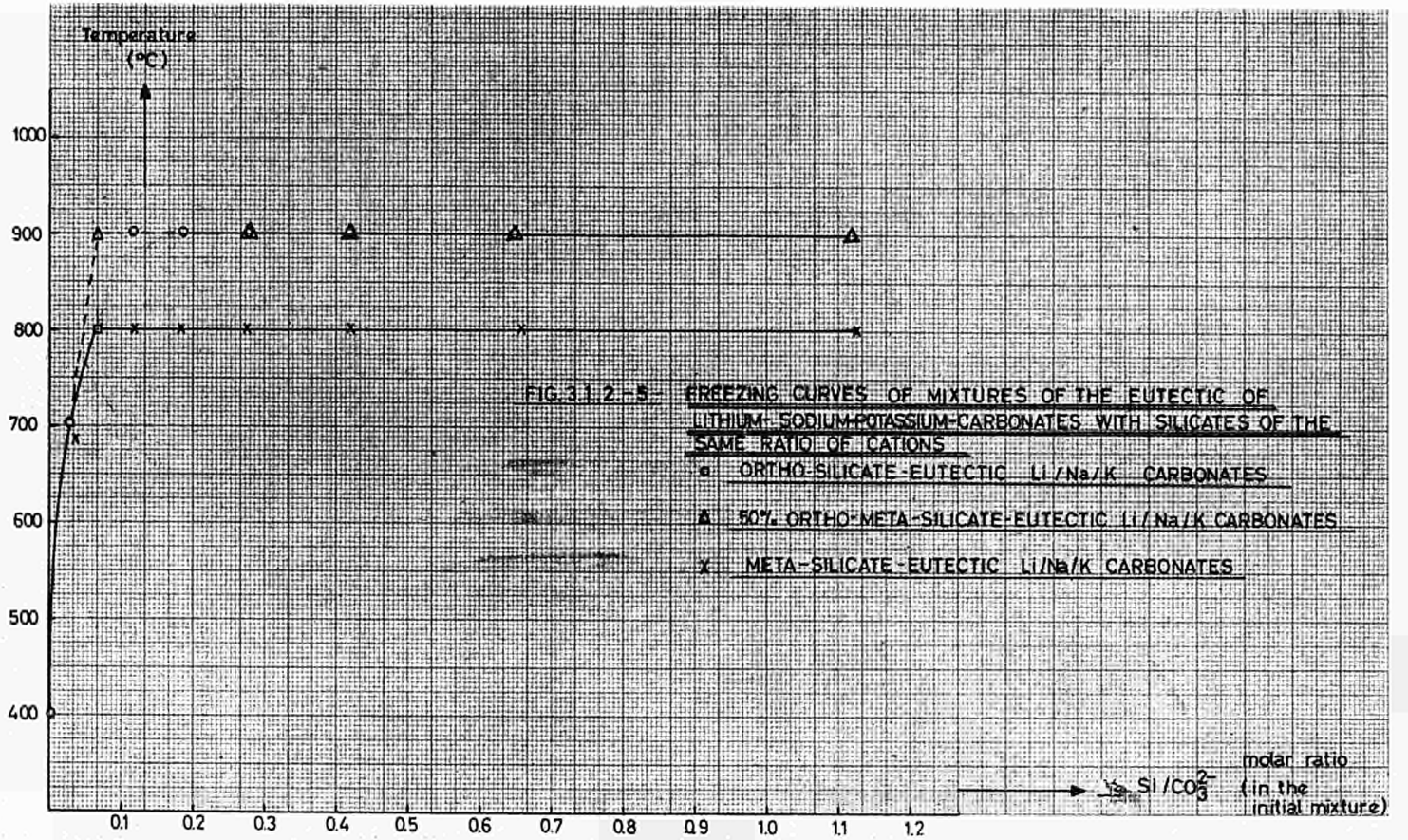
Temperature (°C)
1000
900
800
700
600
500
400
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
molar ratio (in the initial mixture)
Si /CO3 2-
FIG. 3.1.2.-5- FREEZING CURVES OF MIXTURES OF THE EUTECTIC OF LITHIUM-SODIUM-POTASSIUM-CARBONATES WITH SILICATES OF THE SAME RATIO OF CATIONS
o ORTHO-SILICATE-EUTECTIC Li/Na/K CARBONATES
Δ 50% ORTHO-META-SILICATE-EUTECTIC Li/Na/K CARBONATES
x META-SILICATE-EUTECTIC Li/Na/K CARBONATES

FIG. 3.1.2.-5-

The following conclusions can be drawn from the results shown in Figs. 3.1.2.-1, 2, 3, 4- :

(1) the pyrolytic carbon coating burns in oxygen or air in the temperature range 650-1,050° C at a rate increasing with increasing temperature;

(2) the "silicon dicarbide" layer does not react in the temperature range 650-1,050° C to the extent of being shown,
   — either by a complete disintegration of the particles,
   — or even by a detectable weight gain as expected from the stoichiometry of the $SiC_2 + 3O_2 \rightarrow SiO_2 + 2CO_2$ reaction. The fact that no detectable reaction takes place at 650° C agrees with the data for silicon mono-carbide compiled by Lea [5], who concludes, as mentioned before, that its oxidation begins in the neighbourhood of 800° C. The fact that no reaction was detected at 1,000 or 1,050° C may be explained by a very slow diffusion controlled reaction due to the formation of layers of SiO_2 as observed by Dillon [2] and by Jorgensen [3] in the oxidation of carborundum by dry oxygen in the temperature range 900-1,600 °C.

(*b*) Thermolysis in molten alkaline baths.

The results of the first type of experiments described in 3.1.1. (*b*) are summarized in Table 3.1.2.-1-.

As shown, a first approach was made by generating the oxygen in the bath by the thermolysis of an alkaline nitrate. The conclusions from this first approach are that under the particular conditions of dissolved and atmospheric gases (O, O_2, NO_2, NO), the minimum feasible temperature for reaction is :

— 500° C in the NaOH bath, and,

— 600° C in the alkaline carbonates bath.

These conclusions appear to be valid when the oxidant is supplied to the melt as gaseous molecular oxygen, air or nitrogen dioxide.

A second factor deciding the minimum temperature of the process for attacking silicon dicarbide coatings by alkaline melts lies in the influence of the temperature on the solubility of the silicates in the bath.

Figs. 3.1.2.-5, 6, 7- give the results obtained for the five different types of baths prepared in the way described in 3.1.1.(*b*).

From the data shown in those figures, the following comments can be made about the minimum limit for the temperature of the silicon dicarbide decoating process in a bath consisting initially of the eutectic mixture of lithium, sodium and potassium carbonates :

— the silicon dissolved as orthosilicate of the same cations in the same ratio could lead to Si/CO_3^{2-} ratios (1) above 0.1 only at 900° C;

— the silicon dissolved as metasilicate of the same ratio of cations could lead to Si/CO_3^{2-} ratios (1) above 0.1 at 800° C;

(1) Referred to initial CO_3^{2-}.

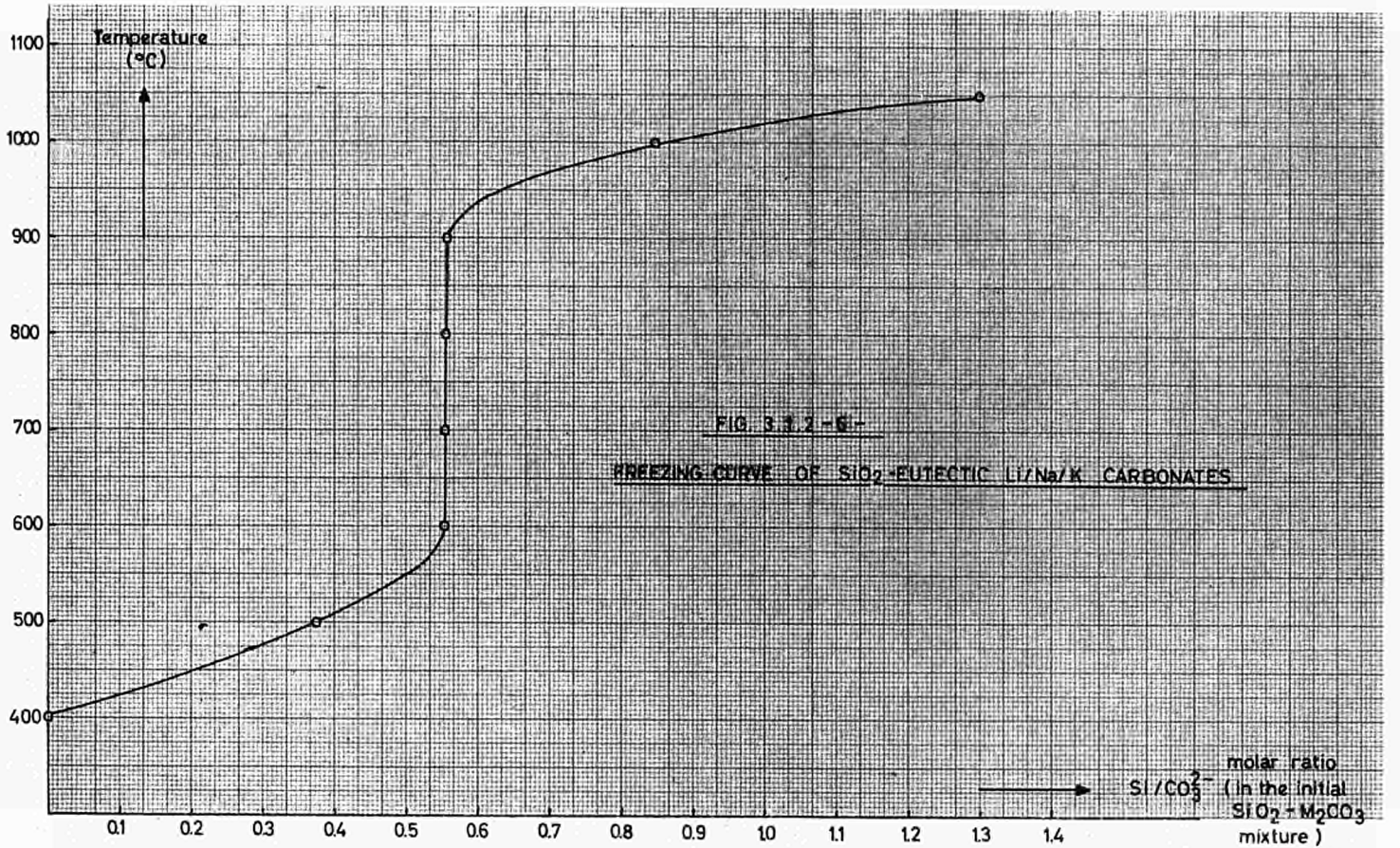
Temperature (°C)
1100
1000
900
800
700
600
500
400
FIG. 3.1.2.-6-
FREEZING CURVE OF SiO_2-EUTECTIC Li/Na/K CARBONATES
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
1.3
1.4
Si/CO_3^{2-} molar ratio (in the initial $SiO_2 - M_2CO_3$ mixture)

FIG. 3,1.2.-6-

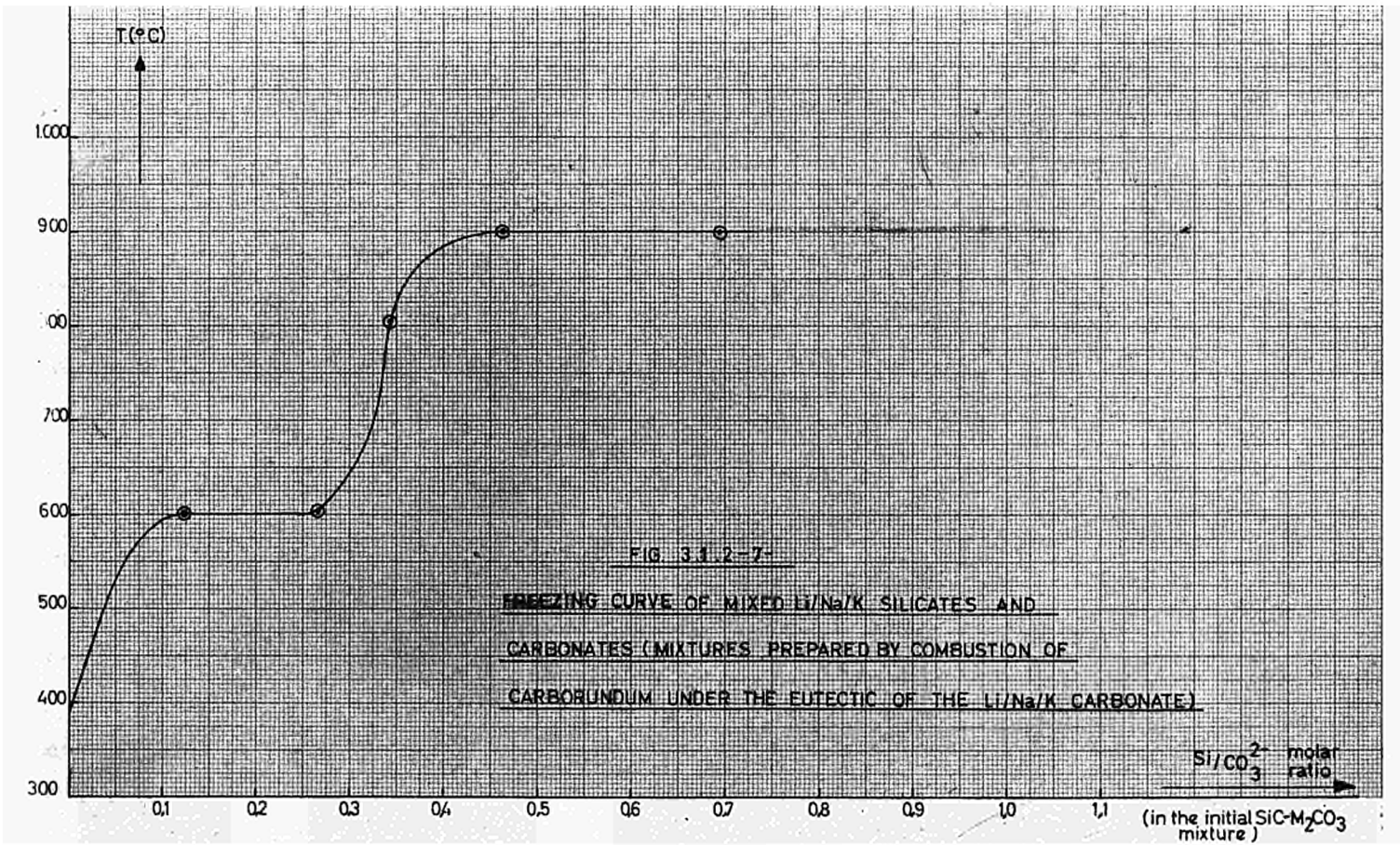
T(°C)
1.000
900
00
700
600
500
400
300
0,1
0,2
0,3
0,4
0,5
0,6
0,7
0,8
0,9
1,0
1,1
Si/CO3 2- molar ratio
(in the initial SiC-M2CO3 mixture)
FIG. 3.1.2.-7-
FREEZING CURVE OF MIXED Li/Na/K SILICATES AND
CARBONATES (MIXTURES PREPARED BY COMBUSTION OF
CARBORUNDUM UNDER THE EUTECTIC OF THE Li/Na/K CARBONATE)

FIG. 3.1.2.-7-

— the silicon dissolved as an unknown mixture of the different silicates resulting from the dissolution of SiO_2 in the eutectic of the three carbonates, could lead to Si/CO_3^{2-} ratios up to 0.55 below 600° C, but to dissolve more silicon, to give a higher value, a temperature of 900° C was required;
— the silicon dissolved as an unknown mixture of silicates resulting from the combustion-dissolution of silicon carbide under the eutectic of the three carbonates could lead to a Si/CO_3^{2-} ratios (1) of 0.25 at 600° C, but to lead to Si/CO_3^{2-} ratios (1) above 0.4 a temperature of 900° C was required.

These last values have been accepted as corresponding most closely to those of process conditions and have been used as the basis for discussion of silicon dicarbide decoating processes under the eutectic of the carbonates of lithium, sodium and potassium.

All the ternary mixtures of alkali hydroxides, carbonates and silicates prepared in the way described in 3.1.1. (*b*) gave a solid phase unless a temperature of 900° C was reached. The results show that, for a Si/CO_3^{2-} ratio 0.5, which is the minimum possible in a combustion-dissolution reaction of SiC_2 under any alkali hydroxide, a temperature of 900° C will be required to keep one single phase in the bath.

(*c*) Effect of induction or dielectric heating.

From the very different nature of the core, formed by the so-called metallic carbides (uranium dicarbide-thorium dicarbide or uranium monocarbide-zirconium monocarbide) and the coating layers (carbon, the non metallic carbide silicon dicarbide) and the small thickness of those layers, one might expect :
— substantial difference in certain physical properties of these materials, i.e. electrical and thermal conductivity (silicon monocarbide is a semi-conductor, while graphite is known to be a good conductor), thermal expansion coefficients; and as a consequence,
— feasibility of rupture of the silicon dicarbide layer by submitting the particles to high frequency electrical fields.

None of the high-frequency generators tested had a substantial influence on the particles : the micrographs before and after the tests appeared quite alike (Fig. 3.1.2.-8-). Only in accidental cases, in which the dielectric field initiated an arc striking the spheres, was a deformation of the particles observed but there was still no rupture.

These experiments were clearly negative, although the limitations in the frequency of the field, the power of the gencrator and the arrangement of the experiments must be pointed out. Nothing can be said about the effect of higher power induction heating, or higher frequency dielectric heat on both the carbon and the silicon dicarbide layers.

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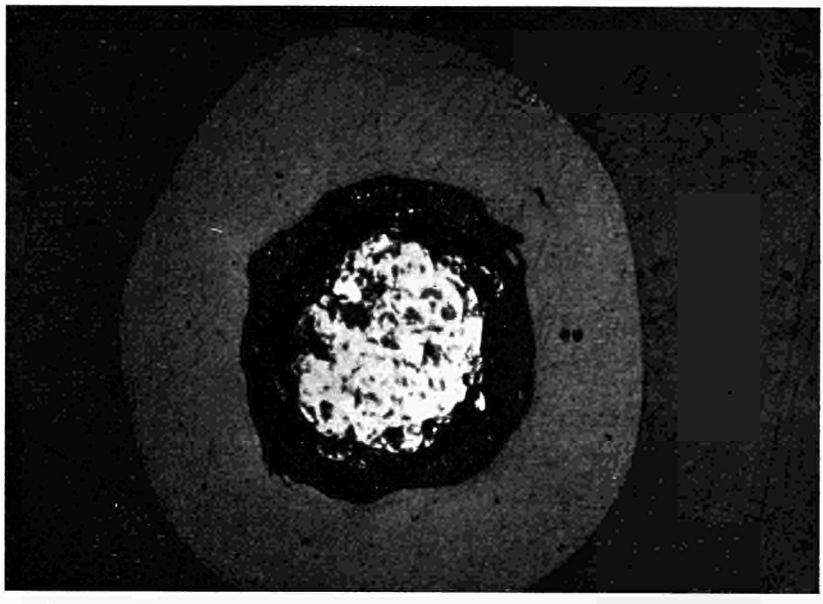
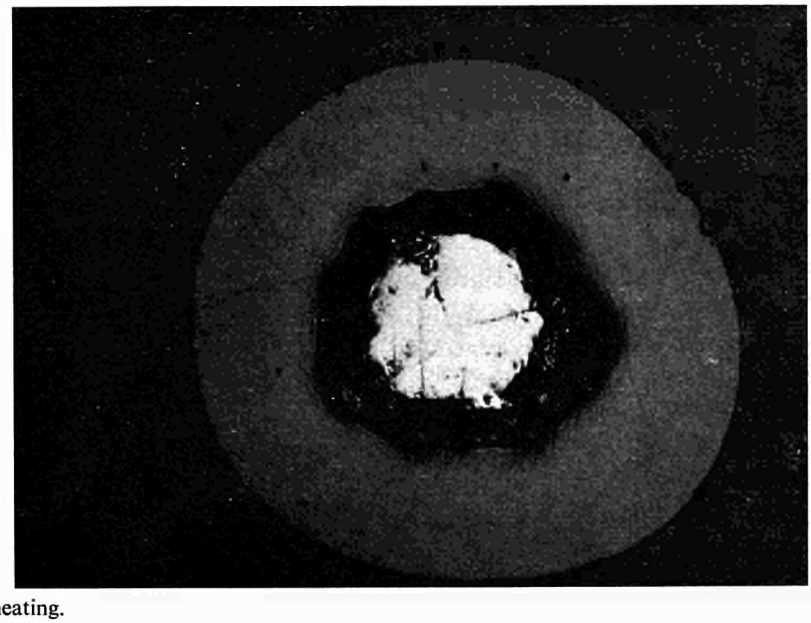
(1) Referred to initial CO_3^{2-}.



Before heating.



After heating.

FIG. 3.1.2.-8- — Micrographs of type A particles before and after submission to induction heating.

(*d*) Other experiments.

All the reactivity tests with aqueous reagents described on 3.1.1. (*d*) were negative.

3.1.3. — *Conclusions.*

Two conclusions can be drawn from the results presented in this section :

1. — *A silicon dicarbide decoating process* appears *feasible*, based on its reaction with air, oxygen or nitrogen dioxide, at a temperature of *600° C* under molten baths consisting initially of the eutectic of lithium, sodium and potassium carbonates.

   The *maximum possible silicate concentration* in the melt corresponds to a Si/ alkaline cations ratio of 1/8, i.e., to a ratio of Si/CO_3^{2-} in the initial bath of 1/4.

   The *temperature of reaction* must be 600° C from the beginning of the attack.

   The *reaction rate* seems but is not quantitatively proved to be higher when hydroxides are present in the bath.

2. — The *removal* of the *external pyrolytic carbon layer* appears *feasible at* 600° C :
   — either by "dry combustion",
   — or, combustion under the same melt used for the silicon dicarbide decoating.

3.2. — THE GRAPHITE.

The study of the rate of the reaction of the graphite, used in the fabrication of carbide-graphite fuel elements, with air, oxygen or nitrogen dioxide,

— either in a "dry" combustion process,
— or in a process of combustion under molten alkaline baths, appeared necessary, considering :
   — the possible existence of fuel elements in which an easy mechanical separation of the coated particles from the graphite matrix would not be feasible (i.e. "dry" combustion of the graphite required),
   — the results of the experiments on the pyrolytic carbon-silicon dicarbide process (i.e. combustion under alkaline melts required),
   — the fact that a "dry" combustion of the graphite matrix followed by a combustion of the coated particles in a molten bath will imply an intermediate period in which a combustion of graphite under the melt takes place.

3.2.1. — *Experimental Techniques.*

(*a*) "Dry" combustion with oxygen or nitrogen dioxide.

The rate of reaction of graphite with oxygen or nitrogen dioxide at different temperatures was studied :

— on electrolytical grade graphite cyclindrical compacts, 20 mm diameter and 25 mm long, with a density of 1.65 g/cm^3,
— in the same 78 mm internal diameter cylindrical nickel reactor described in 3.1.1.(*b*)

The reaction was carried out with circulation of the corresponding gas (atmospheric pressure) at a standard 24 l/h flow, measured at room temperature before the gas entered the reactor.

In each experiment, one compact was placed on its side in the centre of the bottom of the reactor and after a time of reaction of 1 hour its weight loss determined. The weight loss per unit of time and unit of initial surface area was arbitrarily defined as initial rate of reaction, which is roughly correct provided that the loss of weight is not so high during the time fixed that too great a change of the surface area occurs.

(*b*) Combustion under molten alkaline baths.

The same equipment, compacts and experimental arrangement as for the "dry" combustion were used to determine the rate of reaction of the graphite with oxygen and nitrogen dioxide inside a molten bath consisting of the eutectic mixture of lithium, sodium and potassium carbonates.

After cooling the bath down to room temperature and dissolution of the carbonates with dilute nitric acid, the weight loss of the graphite was determined and the initial rate of reaction determined as for the "dry" combustion process.

3.2.2. — *Results.*

(*a*) "Dry" combustion with oxygen or nitrogen dioxide.

The initial rates of reaction of graphite with O_2 and with NO_2 in the temperature range 400-900° C are given in Fig. 3.2.2.-1-.

Features to be noted in the graphs of "dry" combustion with O_2 and with NO_2 are :

— practically no reaction with O_2 at 400° C, while the reaction rate with NO_2, is already fairly high at that temperature. This might be explained by the fact that the reaction defining the process rate at that temperature is the reaction of carbon with elemental oxygen and the partial pressure of elemental oxygen is very much higher from the thermolysis of nitrogen dioxide than from the thermolysis of molecular oxygen at 400° C : the thermolysis of NO_2 to NO and elemental oxygen is known to be 25 % displaced toward the decomposition products at 300° C, and 75 % at 500° C [11] while the thermolysis of molecular oxygen to elemental oxygen requires 1,500° C to reach 1 % decomposition [11];

— a higher rate of the reaction with nitrogen dioxide up to at least 500° C and a higher rate of the oxygen reaction above at least 600° C. For this the following explanation might be valid : above the temperatures 500-600° C the rate of the reaction with molecular oxygen becomes competitive with the rate of the reaction with elemental oxygen, and the partial pressure of molecular oxygen is higher when supplied as such than when fed as nitrogen dioxide on the same volume basis;

— a maximum for the rate of both reactions at the same temperature, around 700° C, in agreement with the reported range of values (670-720° C) for the ignition point of the graphite,

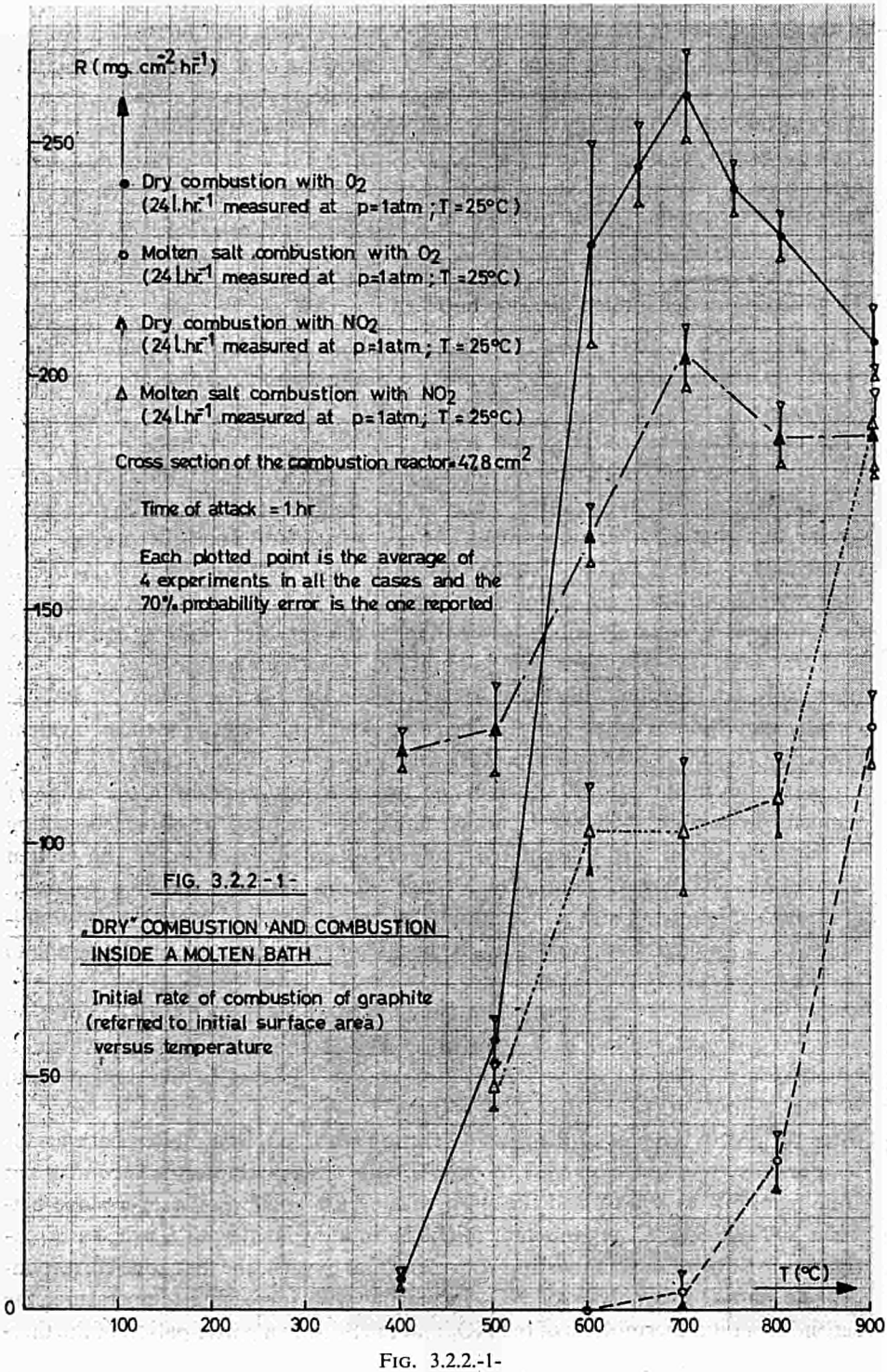

R (mg. cm⁻². hr⁻¹)
250
200
150
100
50
0
● Dry combustion with O2 (24 l.hr⁻¹ measured at p=1atm ; T=25°C)
o Molten salt combustion with O2 (24 l.hr⁻¹ measured at p=1atm ; T=25°C)
▲ Dry combustion with NO2 (24 l.hr⁻¹ measured at p=1atm ; T=25°C)
△ Molten salt combustion with NO2 (24 l.hr⁻¹ measured at p=1atm ; T=25°C)
Cross section of the combustion reactor=47.8 cm²
Time of attack = 1 hr
Each plotted point is the average of 4 experiments in all the cases and the 70% probability error is the one reported
FIG. 3.2.2-1-
„DRY" COMBUSTION AND COMBUSTION INSIDE A MOLTEN BATH
Initial rate of combustion of graphite (referred to initial surface area) versus temperature
T (°C)
100
200
300
400
500
600
700
800
900

FIG. 3.2.2.-1-

— a steeper increase of the rate of the reaction with oxygen with increasing temperature in the temperature range 400-700° C, meaning that the overall reaction of the combustion of graphite with nitrogen dioxide appears as a reaction requiring a lower activation energy than that with oxygen for which 20 kcal $\times$ mol^{-1} have been estimated (as drawn from Fig. 3.2.2.-2- where the reaction rate data have been corrected for the specific surface of the compacts and are given as the fraction of graphite reacted per unit of time versus the reciprocal of the absolute temperature).

(*b*) Combustion under molten alkaline baths.

The Fig. 3.2.2.-1- also gives the initial rates of reaction of graphite with O_2 and with NO_2 inside the molten eutectic mixture of the carbonates of lithium, sodium and potassium.

Features to be noted in the graphs are :

— the rates of reaction, both with O_2 and with NO_2, are lower than those of the corresponding reactions under "dry" combustion conditions. This can be explained by the fact that if other phenomena do not greatly influence the process rate (i.e. catalytic effects of the alkaline cations of the melt), the reaction between the graphite and the gases dissolved in the bath is slower because the concentration of gases is certainly much lower than in the gaseous phase in equilibrium with the melt, owing to their low solubility at these temperatures;

— practically no reaction with O_2 at 700° C, while the reaction rate with NO_2 is already quite important at 500° C. The same explanation given for the equivalent effect in the "dry" combustion process might be given for this case;

— no maximum in the rate of either of the two reactions appears in the range of temperature studied, 400-900° C, which might be explained by two facts, acting in the same sense : (i) none of the reactions of combustion inside the molten bath in the 400-900° C temperature range attains the rate required to reach ignition; (ii) ignition point is not attained because the good heat transfer conditions in a molten bath avoid local peaks of temperature on the graphite surface;

— a somewhat steeper increase with increasing temperature of the rate of combustion with oxygen in a molten bath than in a "dry" combustion process, which, if reliable, might mean a higher "activation energy" process in the molten bath, 47 kcal $\times$ mol^{-1} (Fig. 3.2.2.-2-);

— three regions in the plot of the rate of reaction with NO_2 in a molten bath versus temperature, the first from 500 to 600° C with a steep slope, the second a flat zone from 600 to 800°C, and the third with even a somewhat steeper slope between 800 and 900° C, regions that might be in agreement with a process essentially consisting of the combustion with elemental oxygen, the first zone corresponding to partial thermolysis of NO_2 increasing with increasing temperature, the second to a total thermolysis of the NO_2 and the third to a participation of the thermolysis of the NO ($2NO \rightarrow N_2O + O$) increasing with increasing temperature.

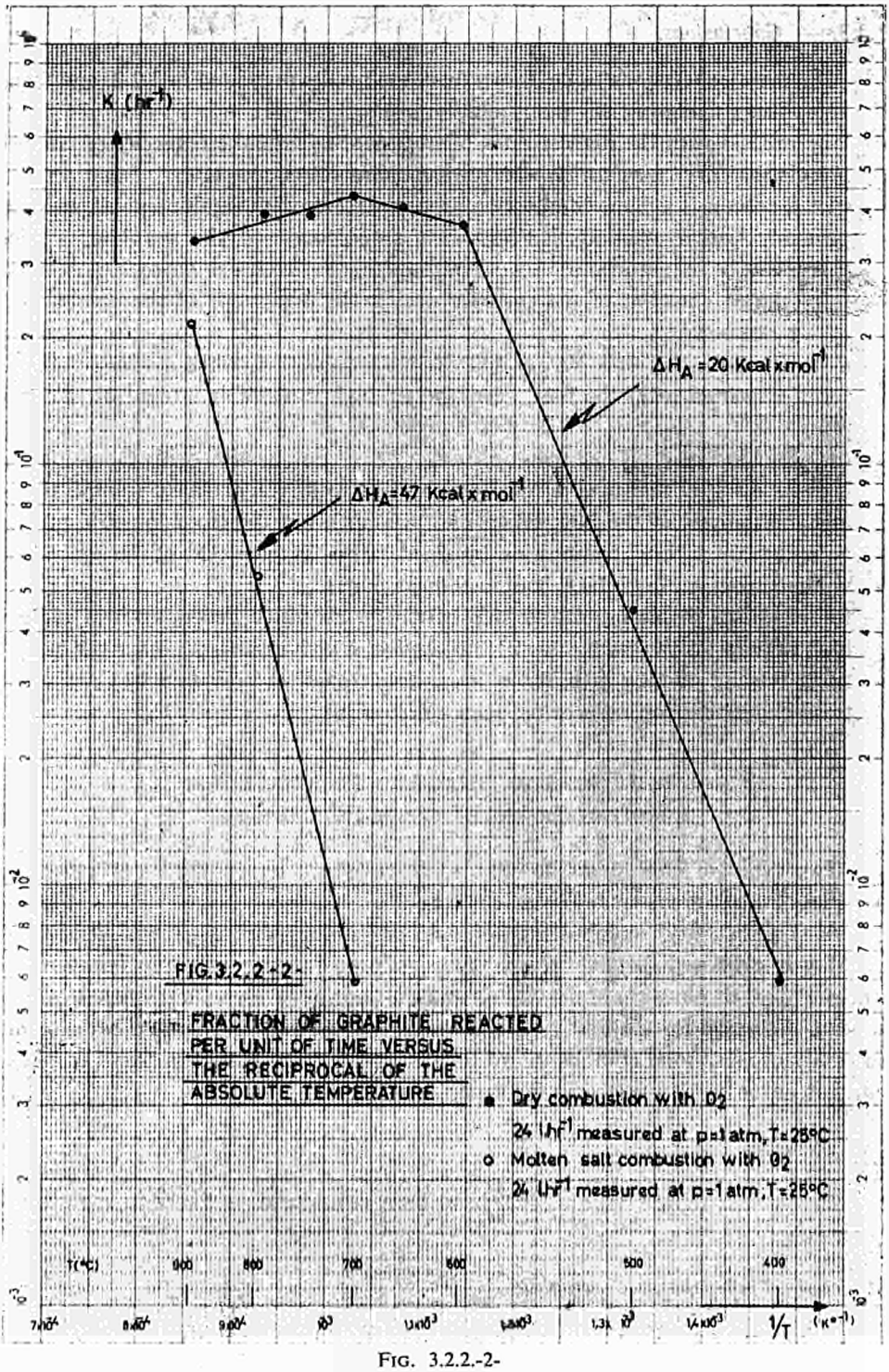

K (hr⁻¹)
ΔH_A=20 Kcal x mol⁻¹
ΔH_A=47 Kcal x mol⁻¹
FIG. 3.2.2.-2-
FRACTION OF GRAPHITE REACTED PER UNIT OF TIME VERSUS THE RECIPROCAL OF THE ABSOLUTE TEMPERATURE
● Dry combustion with O2
24 l/hr⁻¹ measured at p=1 atm, T=25°C
○ Molten salt combustion with O2
24 l/hr⁻¹ measured at p=1 atm, T=25°C
T(°C)
1/T (K⁻¹)

FIG. 3.2.2.-2-

3.2.3. — *Conclusions.*

The main conclusions to be drawn from the results presented in 3.2.2. and from the hypotheses made in their interpretation are :

— the process with the highest rates of treatment of graphite (*capacity of production*) is the "*dry*" *combustion with oxygen above* 700° C;
— such a process is a flame combustion process, i.e. above the ignition point, and will probably lead to uncontrolled *high local temperatures* in the graphite, a fact that might be of importance because of the *refractory nature of the oxides* obtained from the combustion of the carbides during the final steps of the combustion of the carbon;
— *a "dry" combustion with NO_2 at* 400° C is a process that, while *avoiding* the *high* local *temperatures* in the fuel, keeps rather high rates of treatment of the graphite;
— the stoichiometric off-gas volume from a "dry" combustion with O_2 at 700° C is about 40 % of that from a "dry" combustion with NO_2 at 500° C (temperatures and thermolysis of the NO_2 taken into account), but the stoichiometric off-gas volume from a "dry" combustion by air at 700° C is twice that of the equivalent volume from a "dry" NO_2 combustion at 500° C;
— for processes involving a later step of silicon dicarbide decoating in a molten bath at 600° C, *the combustion* of the *residual graphite under the melt* will necessitate the use of NO_2 in order *to avoid temperature as high* as 900° C, required to reach combustion rates of the same order of magnitude with O_2.

4. — Conceptual Head-End Flowsheets for European Graphite-Carbide Fuel Elements

Two types of conceptual flowsheets are envisaged depending on whether the particles of the core are coated by silicon carbide or not.

A straight "dry" combustion followed by a fluoride catalysed nitric acid leaching is proposed for the second type, while an intermediate step between the straight "dry" combustion of the carbon and the fluoride catalysed nitric acid leaching appears necessary for the first type. This step, combustion in a molten bath, seems necessary because total combustion of the silicon carbide coatings in "dry" combustion has never been observed in the experiments reported here.

4.1. — "Schulten" type fuel.

Two models of this type of fuel have been considered as likely to require reprocessing.

Model A : "non-separable" graphite.

This consists of 6 cm diameter graphite spheres in which are dispersed spherical particles of 500 μ external diameter. Each particle consists of a 200-300 μ diameter

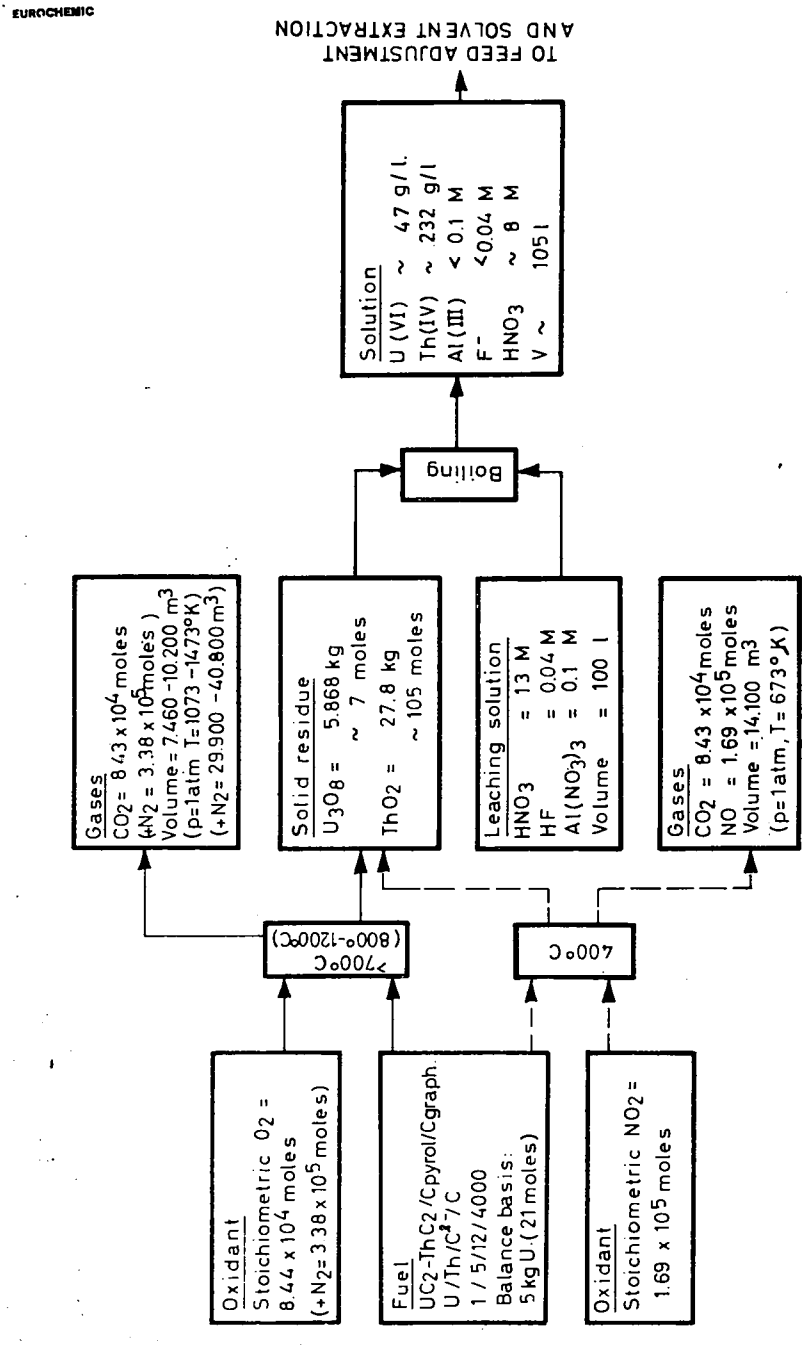


Fig. 4.1-1 – "SCHULTEN"
NON SEPARABLE GRAPHITE

TABLE 4. 1. — Processes for "Schulten" and "Dragon" type fuels

Characteristics of the Process	"Schulten" = model A		"Schulten" = model B		"Dragon"	
	Combustion with		Combustion with		Combustion with	
	O_2	NO_2	O_2	NO_2	O_2	NO_2
1) Semicontinuous combustion in a burner critically safe by geometry	Multitubular reactor consisting of 12 cm diameter cylinders (or slab type)					
Temperature of combustion . . .	>700° C (800-1,200° C)	400° C	>700° C (800-1,200° C)	400° C	>700° C (800-1,200° C)	400° C
Expected (*) rate of combustion (referred to 1 g of uranium)	~40 g carbon/h	~20 g carbon/h	~300 g carbon/h	~150 g carbon/h	~300 g carbon/h	~150 g carbon/h
Required time for combustion of the carbon associated with 5 kg of uranium	~6 h	~12 h	~6 min	~12 min	~6 min	~12 min

2) Semicontinuous combustion in molten bath	Not required				In a separate burner critically safe by geometry or in the burner used for the graphite combustion, if a constructional material suitable for both operations could be found.
3) Semicontinuous aqueous leaching with boiling 13 *M* HNO_3-0.04 *M* HF-0.1 *M* $Al(NO_3)_3$	In a separate reaction vessel critically safe by geometry, or in the graphite burner itself, if a compromise for refractory and corrosion resistance properties can be achieved in the constructional material.				In a separate reaction vessel critically safe by geometry to which the melt is transferred.
Expected average dissolution rate(**) (referred to 1 g mixed oxide) . . .	50-100 mg/h	No data	50-100 mg/h	No data	No data
Required time for the dissolution of 1 batch containing 5 kg of uranium	>15 h	No data	>15 h	No data	No data
Volume of solution (referred to 5 kg of uranium)	~100 l		~400 l		~1.700 l
HNO_3 concentration in the solution	~8 *M*		~8 *M*		~2 *M*

(*) Estimated from the initial combustion rates given in Fig. 3.2.2. 1 and the initial and final geometrical surfaces of the elements.
(**) Roughly estimated from reference 32.

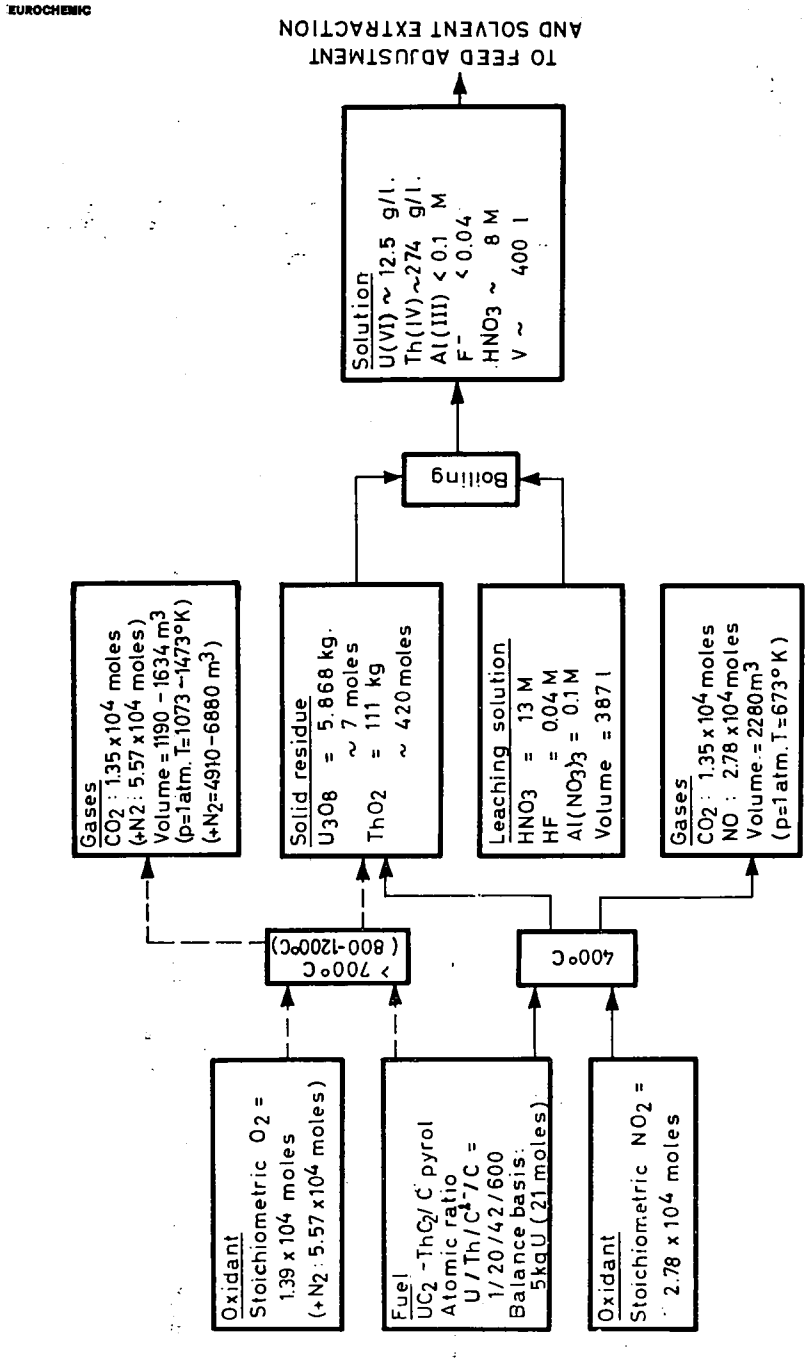


Fig. 4.1-2- "SCHULTEN" SEPARABLE GRAPHITE

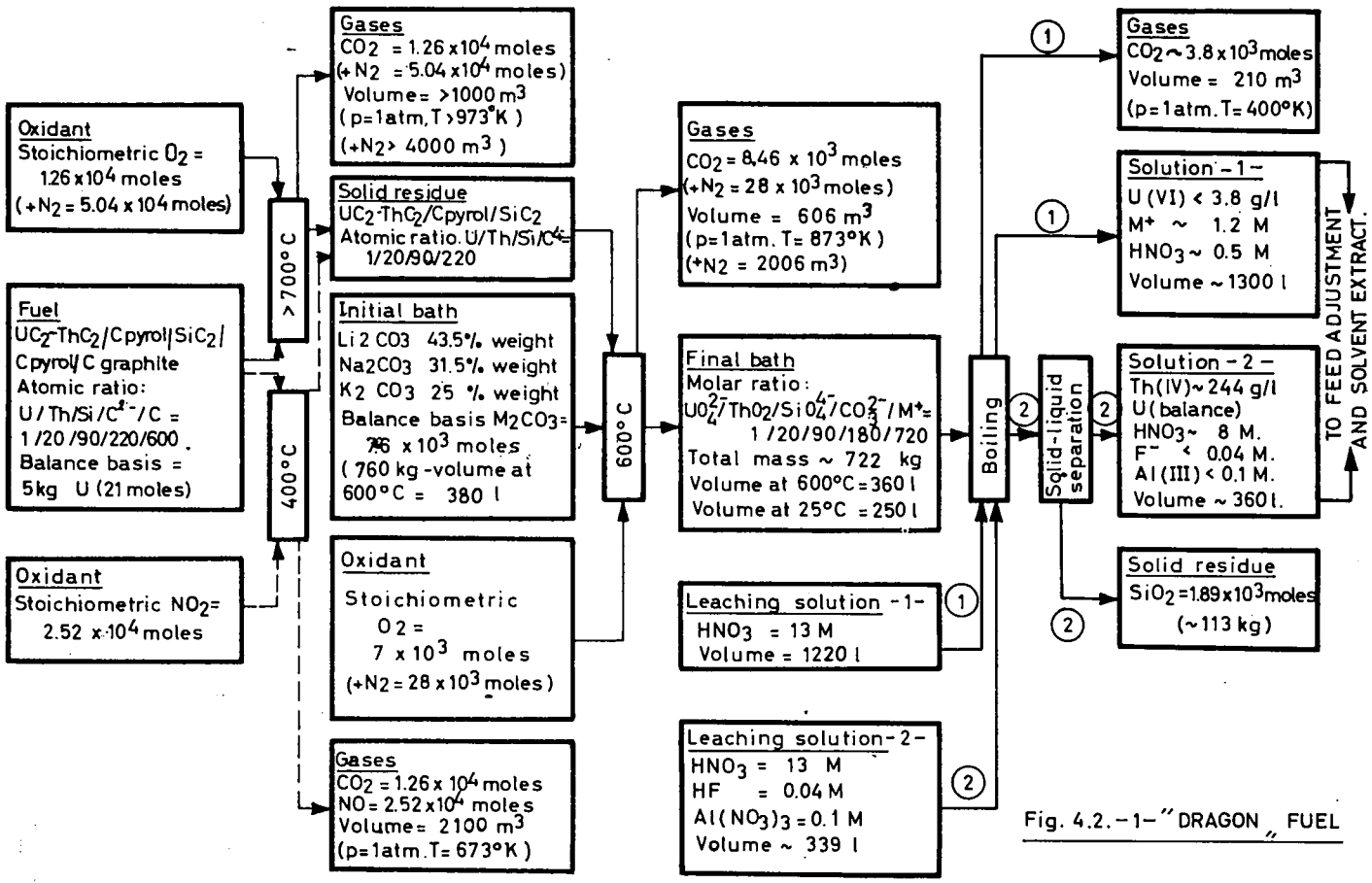


Fig. 4.2.-1-"DRAGON" FUEL

ThC_2-UC_2 sphere with a Th/U ratio of 5, coated with a 30 μ thick pyrolytic carbon layer of 1.5 — 1.6 g/cc density and a second 90 μ thick pyrolytic carbon layer of 1.9 — 2.0 g/cc density. Each sphere contains 200 g of graphite and 1 g of uranium-235. This model would be treated in this form, the particles being inseparable from the graphite matrix after irradiation. The atomic ratio of carbon/uranium in the fuel to be processed will consequently be about 4,000.

Model B : "separable" graphite.

The coated particles, similar to those described above, but with a Th/U ratio of 20, can be separated from the graphite after irradiation. The atomic ratio of carbon of 20, can be separated from the graphite after irradiation. The atomic ratio of carbon/uranium in the fuel to be processed is consequently lowered to about 600.

The proposed head-end flowsheets for models A and B are given in figures 4.1-1- and -2- respectively.

Table 4. -1- gives the main characteristics of the proposed head-end flowsheets for the two models of "Schulten" fuel.

The main feature to be noted is that it is expected that the time of combustion :

— will play a significant role in the capacity of treatment of model A, and,

— will have a negligible influence on the capacity of treatment of model B.

Therefore,

— a combustion with oxygen (or air) above the ignition temperature of the graphite appears advisable for model A, and,

— a *combustion with nitrogen dioxide* below the ignition temperature of the graphite appears worth studying further for the treatment of model B, because the lower temperature, 400° C, to which the cores of the particles are submitted *might lead to milder aqueous leaching requirements.*

4.2. — "DRAGON" TYPE FUEL.

Two types of fuel elements, coming from different zones of the reactor, have been described in the introduction.

The first type has a uranium dicarbide-thorium dicarbide core and the second a uranium monocarbide-zirconium monocarbide core.

The flowsheet presented in figure 4.2. -1- refers to the first type. It is expected that, if the second type is treated according to the principles of this flowsheet, the zirconium oxide will mainly follow the silicon oxide solid residue, but nothing can be advanced at present about the retention of uranium and thorium in such a solid phase.

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REPROCESSING OF NUCLEAR FUELS BY FLUORIDE VOLATILIZATION WITH SULFUR HEXAFLUORIDE

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ABSTRACT

A new procedure was developed for the reprocessing of nuclear fuels by fluoride volatilization, based on the fact that uranium or other uranium containing fuels can be fluorinated in a single step to uranium hexafluoride with sulfur hexafluoride at temperatures above 800° C. The essential reactions taking place can be represented by the following equations :

$$UO_2 + SF_6 \rightarrow UF_6 + SO_2$$
$$UC_2 + SF_6 + 3O_2 \longrightarrow UF_6 + 2CO_2 + SO_2$$
$$U + SF_6 + O_2 \longrightarrow UF_6 + SO_2$$

Advantageous for the use of sulfur hexafluoride is its property to be non-corrosive up to temperatures of about 500° C.

In order to perform the process, the fuels are first pulverized in a known manner and then fluorinated either with pure sulfur hexafluoride alone or together with oxidation agents like oxygen, air, manganese dioxide or others. The fluorination can be done in two steps. At first the starting material is fluorinated at temperatures between 700° C and 800° C, separating the non volatile uranyl fluoride from the easily volatile fission product fluorides. Afterwards, in the second step the UO_2F_2 is transformed to the easily volatile UF_6 at about 900° C.

For the reprocessing of coated particle containing fuels it has proved appropriate, either to crack and pulverize these particles prior to fluorination or to treat them at temperatures of about 900° C up to 1100° C with oxygen or a mixture of oxygen and sulfur hexafluoride, eventually in the presence of combustion catalysts. The fluorination can be achieved using either the fluidized bed or the conventional rotating oven technique. As contructing material for the ovens, besides others; especially pure alumina has stood the test.

The advantages of the new procedure are not only its simplicity and economy, another important fact is the absolute untoxicity of sulfur hexafluoride. It does not act corrosive on any part of the gas inlet system. Small amounts of fluorine contained in the off-gas stream may be reconverted into sulfur hexafluoride by reaction with sulfur. This adds another advantage of re-using the so formed sulfur hexafluoride. A required high factor of decontamination of the formed uranium hexafluoride may be achieved applying the known absorption-desorption technique on sodium fluoride columns.

1. — INTRODUCTION

A basic question affecting the economics of nuclear power reactor is the selection of a suitable process for the regeneration of nuclear fuels. It is to be expected

that most of the power reactors shall be carbide, metal, or oxide fueled. An unequivocal choice however, as to which type might be the best is as yet impossible. Special attention is required for the reprocessing of nuclear fuels containing coated particles, which are regarded to be the most promising fuel type for high temperature gas-cooled reactors. It should well be suited to reprocessing by volatilization, particularly if a separation of fissile and fertile material is desired.

The reprocessing of the graphite fuel element balls of the Jülich AVR-reactor necessitates either a modification of the known aqueous or non aqueous procedures, or the development of a new process especially suited for the pyrolytic carbon coated fuel particles. For some time we have been investigating in our laboratory the possibility of reprocessing such fuels using a combined combustion-fluorination process.

It is a well established opinion that the fluoride volatilization should be able to compete with, or even to displace some day the aqueous regeneration procedures. This statement is based on the facts that :

(*a*) a higher degree of decontamination can be achieved using smaller volumes;
(*b*) smaller expenditures for installations are necessary due to fewer process steps;
(*c*) one obtains directly-concentrated waste residues;
(*d*) criticality dangers are decreased, since perilous concentration aggregations can hardly occur throughout all process steps, and finally
(*e*) the problem of radiation damage of chemicals is unimportant.

In addition, the volatile uranium hexafluoride end-product, present the advantage of being directly introducible into an isotope enrichment cascade.

The transformation of uranium hexafluoride into the oxide, carbide, or metal presents no insurmountable technical difficulties.

It has been established that fluoride volatilization will be likewise suitable to all three separation problems in reactor technology, namely :

1. the separation of plutonium, natural uranium and fission products,
2. the separation of enriched uranium-235, fission products, and alloying metals, and
3. the separation of thorium, protactinium, and uranium.

2. — The Sulfur Hexafluoride Process

A new procedure was developed for the reprocessing of nuclear fuels by fluoride volatilization, based on the fact that uranium or other uranium containing fuels can be fluorinated in a single step to uranium hexafluoride with sulfur hexafluoride at temperatures above 800° C. The essential reactions taking place can be represented by the following equations :

$$UO_2 + SF_6 \rightarrow UF_6 + SO_2$$
$$UC_2 + SF_6 + 3O_2 \rightarrow UF_6 + 2CO_2 + SO_2$$
$$U + SF_6 + O_2 \rightarrow UF_6 + SO_2$$

Advantageous for the use of sulfur hexafluoride is its property of being non-corrosive up to temperatures of about 500° C.

In order to perform the process, the fuels are first pulverized in a known manner and then fluorinated either with pure sulfur hexafluoride alone, or together with oxidation agents like oxygen, air, manganese dioxide or others. The fluorination can be done in two steps. In the first, the starting material is fluorinated at temperatures between 750° C and 800° C, separating the non-volatile uranyl fluoride from the easily-volatile fission product fluorides. Afterwards in the second step, the UO_2F_2 is transformed to the easily-volatile UF_6 at about 900° C. In case it is unnecessary to get the high degree of decontamination for the uranium hexafluoride obtained hereby, the fluorination reaction can be performed in a single step under suitable conditions at temperatures between 850° C and 900° C. Care has to be taken that the fluorine concentration is high enough in the reaction zone to avoid the production of uranium fluoride intermediates. For the reprocessing of coated particle-containing fuels it has proved appropriate either to crack and pulverize these particles prior to fluorination, or to treat them at temperatures of about 900° C up to 1100° C with oxygen or a mixture of oxygen and sulfur hexafluoride, eventually in the presence of combustion catalysts.

The fluorination can be achieved using either the fluidized bed or the conventional rotating oven technique. The best materials for the ovens appear to be especially pure alumina. The fluidized bed technique presents several advantages for the realization of the process, since the equipment does not require any moving parts.

The reaction zone of the oven must be coated with pure alumina, fluorspar, or other fluorine resistant materials. For agitating the fluidized bed nitrogen or air is used. In this new process, removal of the heat of reaction is no where near as big a problem as in the case of using fluorine gas, since the heat of formation for the fluorination of UO_2 with SF_6, for example, is much smaller than for F_2. The values of ΔH_{298} are -60.96 kcal/mole and -252 kcal/mole, respectively. The question of whether the fluorination with sulfur hexafluoride is also possible in molten salts is not, as yet, completely solved. One of the main difficulties are corrosion problems.

The advantages of this new procedure lie not only in its simplicity and economy, but also in the absolute non-toxicity of sulfur hexafluoride. It does not react corrosively on any part of the gas inlet system. Small amounts of fluorine contained in the off-gas stream may be reconverted into sulfur hexafluoride by reaction with sulfur, thus adding another advantage. A high factor of decontamination of the resulting uranium hexafluoride may be achieved by applying the absorption-desorption technique on sodium fluoride columns.

3. — Flowsheet of the Sulfur Hexafluoride Process

A tentative schematic flowsheet for the processing of uranium-containing fuel with sulfur hexafluoride is shown in Fig. 1.

The nuclear fuel is first chopped and pulverized, then stored in a storage container. The total usable volume of the container must be smaller than would contain

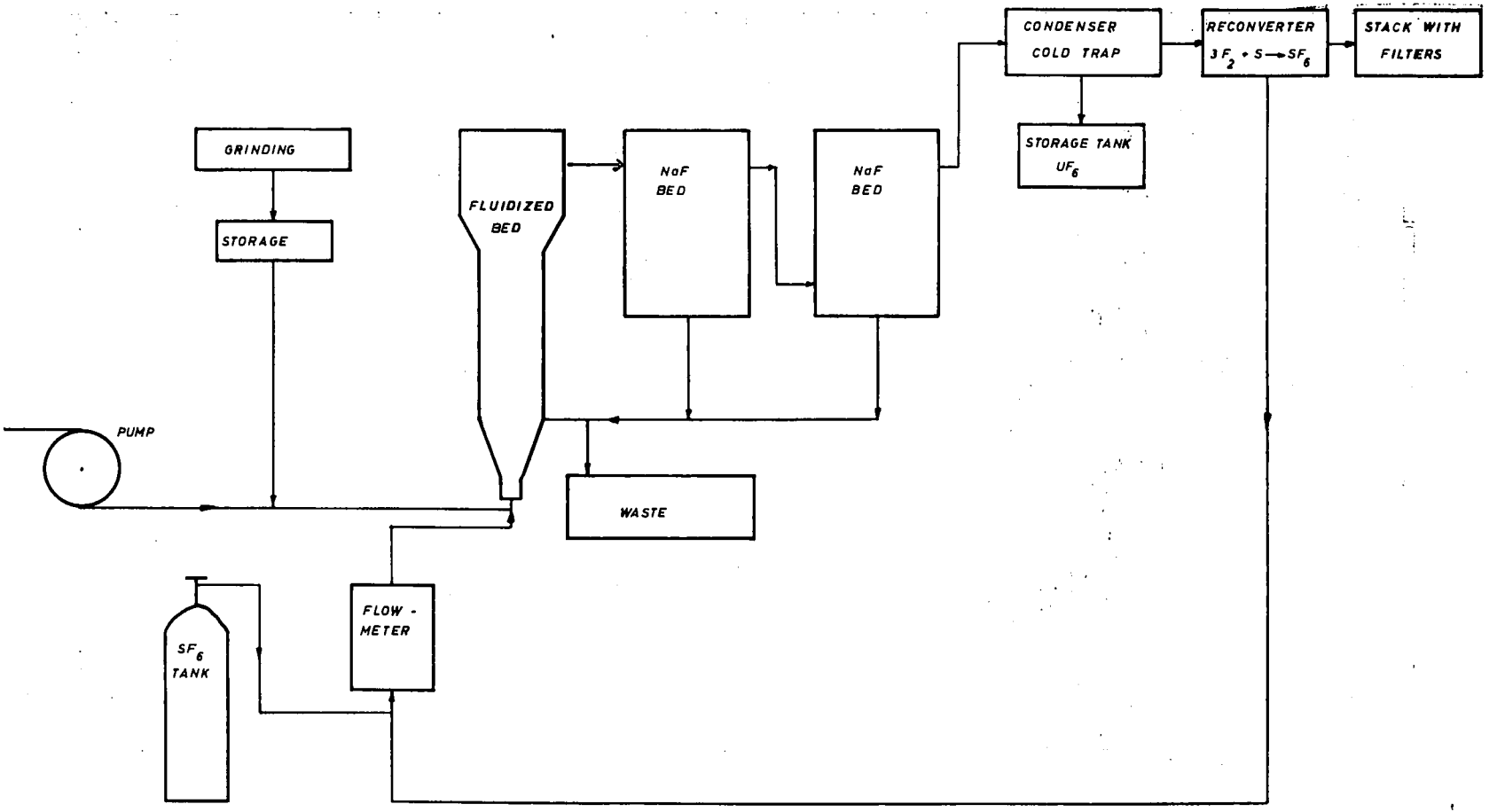

Fig. 1. — Schematic flowsheet of the Sulfur Hexafluoride process.

a critical mass. Using a transport pump and a nitrogen (or air) stream, the pulverized material is fed into the fluidized bed oven together with sulfur hexafluoride. The addition of SF_6 from the storage tank is monitored by observing a flowmeter. The reaction zone of the oven is heated up to about 900° C either by induction or resistance heating.

On top of the furnace the volatile UF_6 thus formed is released and absorbed on a sodium fluoride column. By raising the temperature of the NaF-bed, the UF_6 is desorbed and may be deposited in a cold trap and stored in a special tank. A second sodium fluoride bed is used for additional fission product decontamination. The waste from the process is removed batchwise, and might be poured into cans or long pipes and capped for storage. Fluorine gas which did not react in the fluorinator is reconverted into SF_6 by reaction with sulfur at elevated temperature and may be re-utilized.

4. — Further development

So far, most of the work has been done on a scale of a few grams in cold runs only, with and without the addition of fissium. It was thus possible to gather information on the behaviour of certain important fission products. We are pursuing our initial investigations along three lines :

1. Fluorination of uranium-containing materials with SF_6 in a fluidized bed,
2. Fluorination of uranium-containing materials with SF_6 in a rotating oven, and
3. Fluorination of uranium-containing materials with SF_6 in molten fluoride salts.

The capacity of the different fluorination devices is approximately one hundred grams of uranium. Before starting with highly-irradiated material, we still need much more information from cold runs, especially concerning yields of the different products, reaction mechanisms involved, and construction material problems.

The fluorination with sulfur hexafluoride in a packed bed seems to be more promising than a fluidized one, since here the gas flow is much smaller. This would facilitate the decontamination of the otherwise-arising large amounts of waste gases. Because the heat of formation for SF_6 is markedly less than that of the reaction with fluorine, the risk of caking of the powder is not very big. It can be overcome by mixing the ground uranium-bearing fuel with coarse-grained inert material. The same argument goes for the application of the otherwise-over-used rotating oven. Our present experimental results confirm this supposition. Best results in the fluorination of UF_4 to UF_6 with fluorine were obtained using eutectic mixtures of molten fluoride salts. Whereas the difficult construction material and corrosion problems are fairly well solved using elementary fluorine for the fluorination in molten salts, this is not yet true for SF_6.

Finally, our investigations include the absorption and desorption of UF_6 on NaF-beds and the reconversion of UF_6 to UO_2, using either hydrolytis with water vapor or decomposition in molten salts.

HEAD-END TREATMENT FOR THE REPROCESSING OF HIGH TEMPERATURE CARBIDE-GRAPHITE FUELS

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ABSTRACT

The paper describes an installation for the head-end treatment of carbide-graphite fuel elements. The whole fuel elements are heated in an induction furnace and burned in oxygen and air. The resulting oxides of uranium, thorium and fission products are collected whereas the off-gases containing CO_2, O_2, N_2 and volatile fission products are cleaned in an off-gas treatment system consisting of a scrubber column, an electrostatic filter unit and an absolute filter. The installation has been successfully operated with inactive and slightly active « Dragon-type » fuel elements and is now ready for work with highly irradiated fuel specimens.

1. — INTRODUCTION

In the reprocessing cycle of graphite-carbide fuel for the recovery of uranium and thorium the head-end treatment step needs a special consideration. For obvious reasons, this fuel type has to be treated differently from any fuel studied so far. On the other hand well established techniques may be applied for the decontamination steps and the tail-end treatment of the reprocessing cycle. Therefore no serious problems should be encountered in this part of the treatment. Thus, this paper deals only with the head-end process.

Various methods exist which could be applied to the head-end treatment of graphite-carbide fuel. Most of them have already been tried on a laboratory scale and are listed below :

— Combustion [1], [2], [3], [4].
— Grinding-leaching [1], [3], [5], [6], [7], [14], [15], [16], [17], [18].
— Electrolytic disintegration [1], [3], [8], [9].
— Chemical disintegration [5], [9], [10], [11], [12], [13].
— Volatility processes [5], [11].

2. — SELECTION OF THE HEAD-END PROCESS

The combustion method, which is based on oxidation of the fuel in oxygen at elevated temperatures removes all carbon and leaves a residue of uranium and thorium oxides. These oxides are easily dissolved in a solution of concentrated nitric acid containing fluorides. This method seemed to us to be the most attractive one because of its fundamental simplicity and its straight-forward way to existing solvent

extraction procedures for further reprocessing. Its application, however, involves many engineering problems, particularly the handling of large volumes of highly radioactive exhaust gases and volatile materials. Furthermore it does not attack ceramic coated particles. It could nevertheless have a very useful application for fuel containing silicon carbide coated particles because the volume of the fuel can be reduced considerably by burning the graphite matrix of the fuel elements.

Most of the other processes mentioned above involve a mechanical disintegration of the fuel elements prior to the actual treatment, which seems to us to be a rather difficult operation with highly active materials.

3. — Principle of the combustion method

The basic idea of the process described in this paper is to oxidize the complete fuel elements (i.e. graphite canning plus fuel inserts) or fuel balls in a stream of oxygen and air without previous mechanical disassembly of the elements. The burning of graphite in form of a lump or a thick rod needs a rather high temperature to initiate the reaction, even in an atmosphere of pure oxygen so that an external heat supply must be provided. A normal resistance furnace is not very suitable for this purpose due to the poor and slow control possibilities. Furthermore the high temperature required to reach appreciable burning rates involves construction material problems for the furnace which are not easy to overcome. In order to avoid these difficulties a high frequency induction furnace was developed which does not need much maintenance due to its simplicity. Induction heating is very appropriate because it is easy to control and may be directed to the proper burning zone. Furthermore the graphite canning acts as a susceptor for the high frequency energy, which is very important to reach the high temperatures necessary to decompose coated particle fuel without damaging the furnace mantle.

Once the reaction has started the heat supply may be reduced; the graphite burns to carbon-oxides whereas the uranium and thorium remain as oxides in the furnace. The cooled off-gases need to be treated by a suitable filtering system to remove dust containing uranium and thorium oxides and volatile fission products.

4. — Description of the process

The flow sheet of the head-end treatment is shown in fig. 1. The fuel rod is introduced through the top opening of the furnace and stands in a vertical position on the grate inside the furnace. The lower end of the rod is heated by induction heating until the burning temperature is reached. Oxygen and air are introduced at the lower end of the furnace and the temperature is raised to a proper level by adjusting the power of the high frequency generator. The fuel element starts burning in a small zone and moves down by gravity along the furnace while it burns. A steep cone is formed which stands on the grate and supports the fuel element. The ash, consisting of various oxides of uranium, thorium, fission products, inorganic

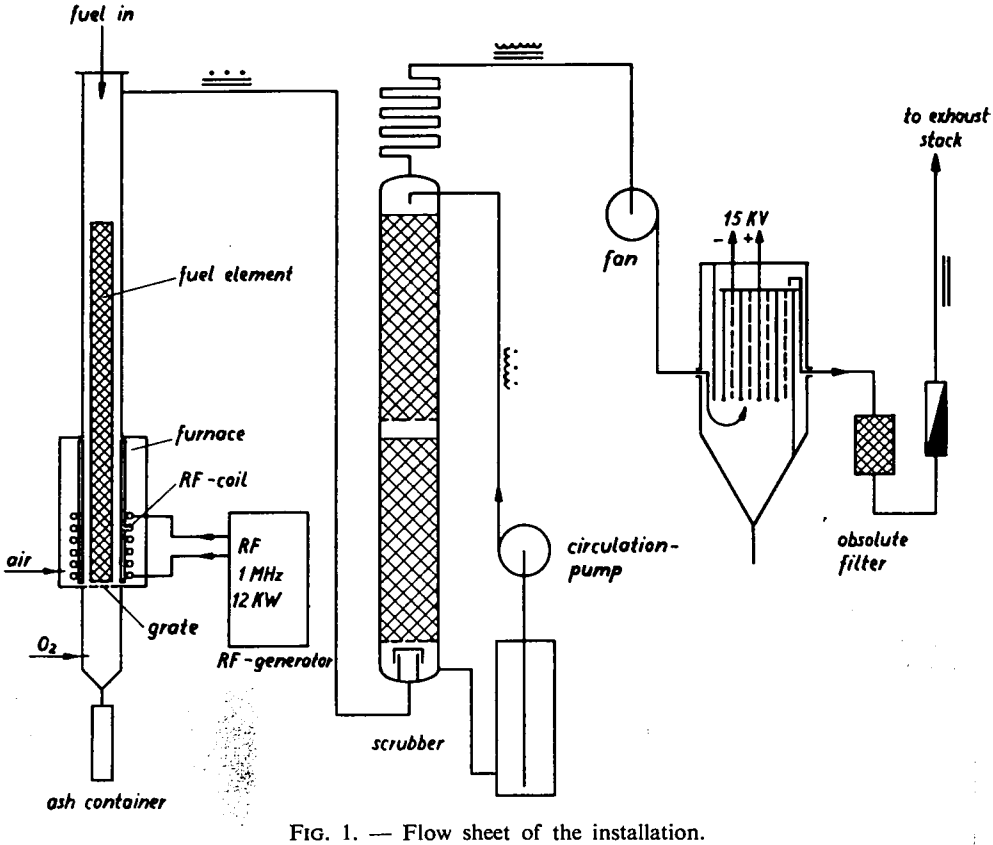

FIG. 1. — Flow sheet of the installation.

coating materials and traces of graphite falls through a valve section into a special, geometrically safe container from where it may be transferred to the dissolver of the reprocessing system. The off-gases consisting of CO_2, O_2, N_2, fission products and traces of the oxides are passed through a scrubber column in which they are washed by a counter currently flowing solution of 5 % sodium bicarbonate. After passing the scrubber unit, the off-gases are forced through an electrostatic filter unit where liquid and solid particles are deposited. From here they flow through an absolute filter unit and are led to the off-gas system of the hot laboratory.

5. — DESCRIPTION OF THE INSTALLATION

The installation may be divided in three main parts : the furnace, the power supply and the off-gas treatment system (fig.2). The furnace and the off-gas handling system are adapted for remote operation and are installed in a 1,000 MeV-C hot cell. The furnace may be disassembled by manipulators. In order to facilitate the manipulator operation it is mounted in a turnable rack, so that it is readily accessible from all sides. The high frequency power supply unit and the power supply of the electrofilter are installed at the outside of the hot cell.

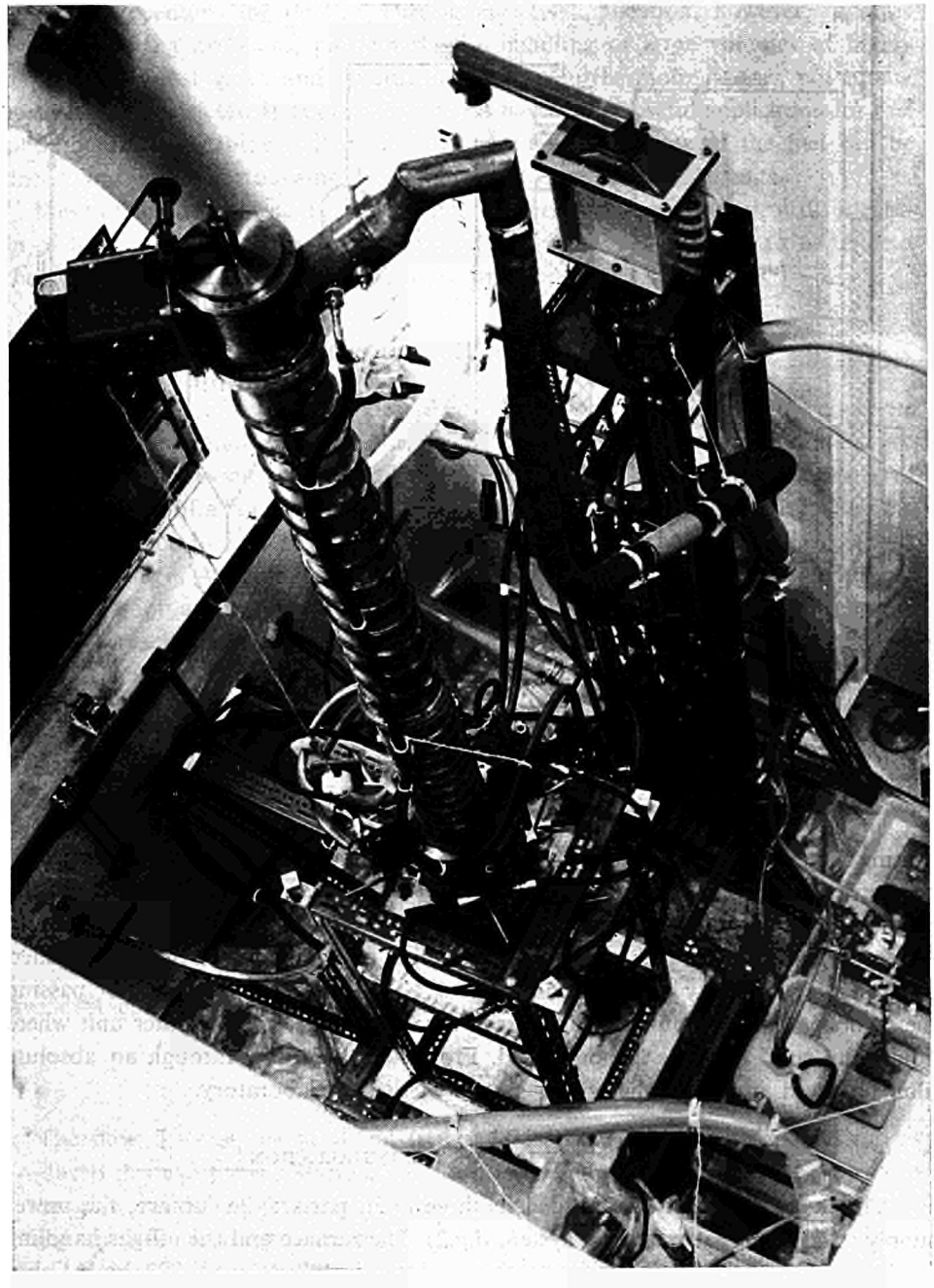

FIG. 2. — View of the installation in the hot-cell. The picture is taken through the open top of the hot-cell. The furnace is in the center of the picture; on its right is the scrubber column with the top-condenser. The thick tubes (neoprene and PVC) belong to the off-gas system, the small black tubes are for cooling-water or hydraulic connections.

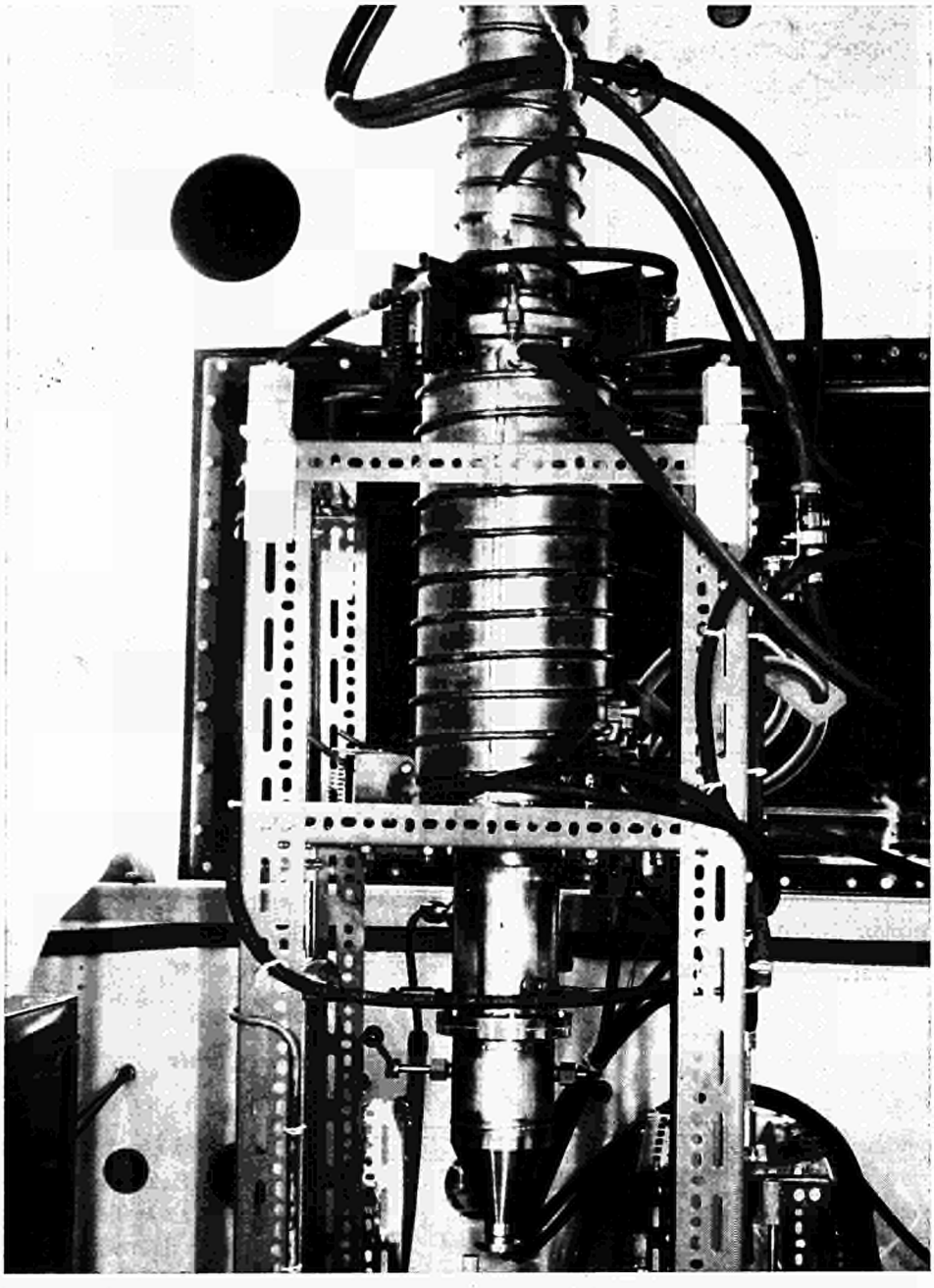

FIG. 3. — Detail of the furnace. The thickest part with the cooling coils contains the high frequency heating system and the MgO furnace tube. Connected with hydraulic, remotely operabable clamps are : — at the top of the furnace the 1,700 mm long stainless steel tube, — at the bottom of the furnace the tube containing grate and cutting device for unburned pieces of fuel. At the lower end the conical part with the ash discharge valve is shown.

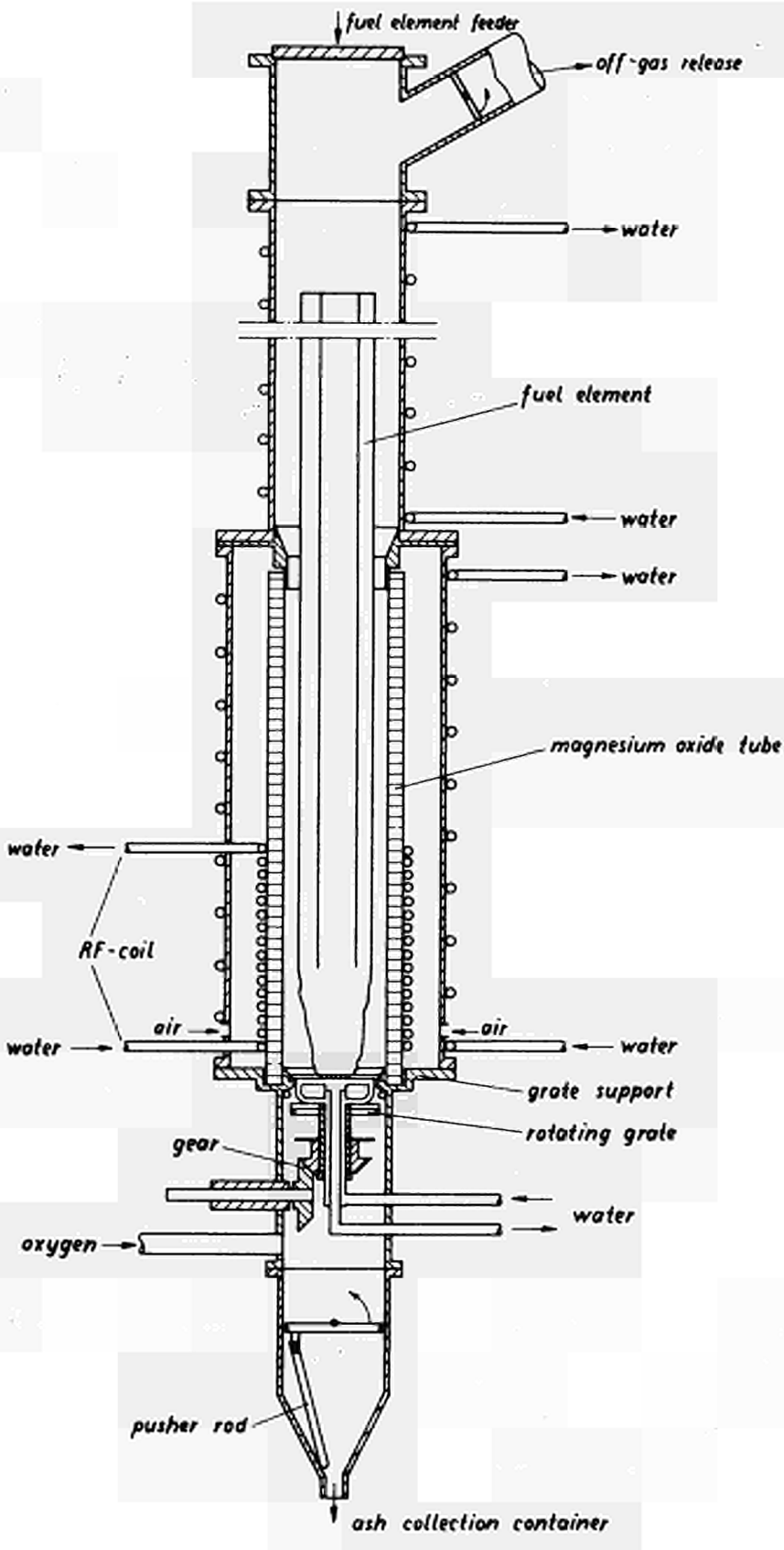

FIG. 4. — Section through the furnace part of the installation.

5.1. — THE FURNACE.

The furnace (fig. 3 + 4) consists of a 200 mm inner diameter vertical stainless steel tube, 500 mm long with a silver soldered cooling coil on the outside. It is equipped with two insulated through-connections for the high frequency supply. Two small tubes for air supply are positioned diametrically at its lower end. The stainless steel tube serves as the containment for the furnace, which consists of a magnesium oxide tube of 100 mm inner diameter, 130 mm outer diameter with the water cooled induction coil around the outside.

A stainless steel tube (130 mm dia, 1,700 mm long) with cooling coils is put on top of the furnace section. A tight connection between both tubes is made by 4 hydraulic clamps which may be disconnected with the aid of the hot cell manipulators to allow remote disassembly of the furnace. The top cover of the furnace may be opened with a motor driven valve to allow the loading of the furnace with fuel. A tube (50 mm dia) at one side of the upper end leads the off-gases to the scrubber unit. Placed below the furnace tube is a section containing a water cooled hexagonal grate and a device which allows the breaking down of bigger fuel parts falling through the grate (fig. 5). The temperature and the flow rate of the cooling water of the grate is measured at the exit. Directly below the cutting device the oxygen inlet tube is placed. The furnace is attached to the grate section by 4 pneumatic clamps and can be removed with the manipulators. A spraying system at the top of the furnace permits rinsing for decontamination purposes. The ashes of the burned fuel and some unburned material fall through the grate to an ash discharge valve (butterfly type) where they may be transferred to the geometrically safe ash container.

5.2. — THE HEATING SYSTEM.

A Philips 12 kW high frequency unit (frequency $\sim$ 1,000 kc) serves as power supply. The high frequency power is led through the cell wall by specially designed lead filled plugs which contain the water cooled copper tubes in a second, steatite isolated tube. The high frequency is not subject to over-heating the plug. The high frequency connections may be disconnected with the manipulators inside the hot cell.

5.3. — THE OFF-GAS TREATMENT SYSTEM.

The off-gas treatment system consists of a sodium bicarbonate *scrubber column*, a blower, an electrostatic filter unit and an absolute filter (fig. 1 + 2).

The off-gases which are cooled by passing the upper parts of the furnace are directed through flexible tubes to the bottom of a sodium bicarbonate scrubber column with the aid of a blower, which is positioned between the scrubber unit and the electro-filter, producing a negative pressure within the furnace and the scrubber unit but a slight over pressure in the rest of the system. The counter current scrubber unit consists of a glass column (100 mm i.d.) with two stainless steel wire gauze Raschig ring filled sections of 800 mm height each. The Raschig rings are supported

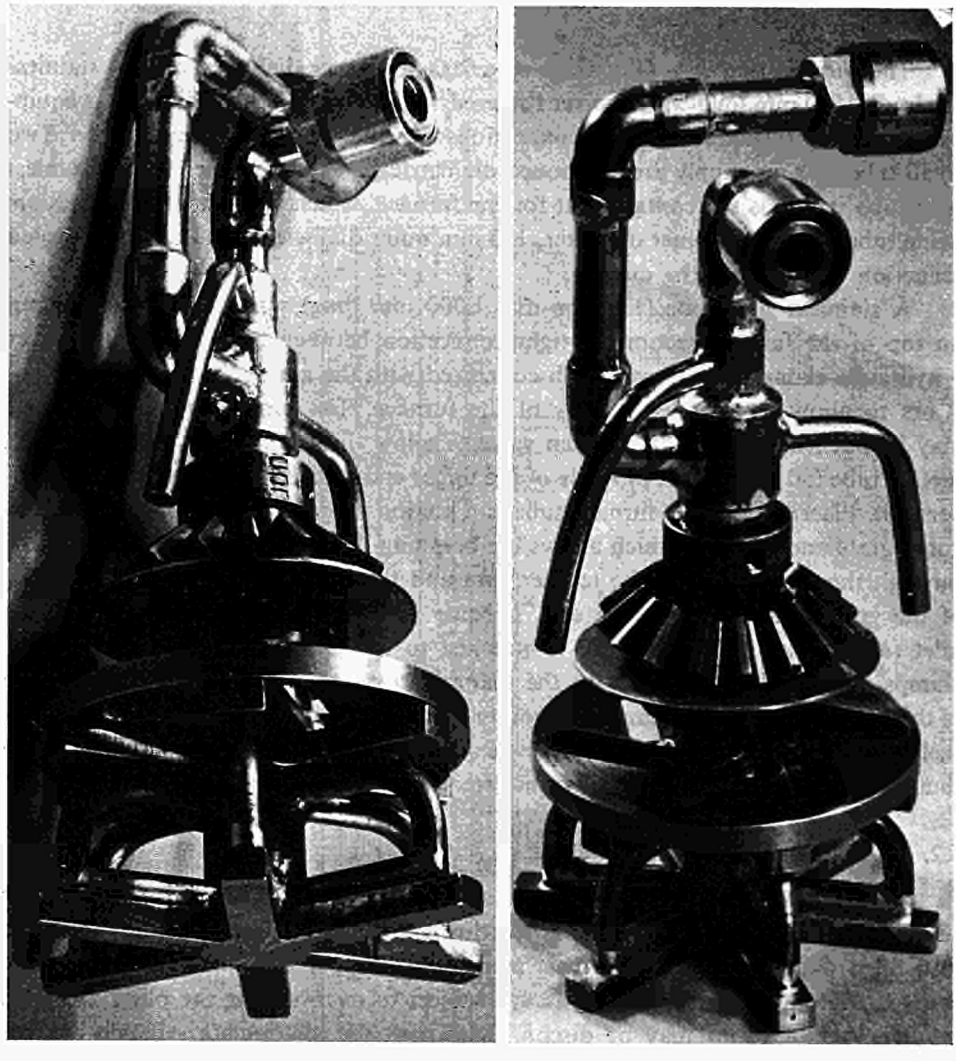

FIG. 5. — The two pictures show the grate with its cooling tubes and the cutting device for bigger, unburned fuel pieces. The gear wheel seen in the pictures activates the cutting device.

by sieve plates and are percolated from top to bottom with a 5 per cent sodium bicarbonate solution which in our experiments was circulated at a rate of about 2 liters per minute. The off-gases are passed through this column in counter current leaving most of the activity. A condenser at the top of the scrubber unit prevents losses of the solution by evaporation and dries the off-gases to a quality suitable for the proper operation of the electrofilter. The two stage centrifugal blower vents the off-gases at a rate of about 2,000 1/h through the system.

The off-gases are blown from the blower to a modified two stage Honeywell *electrofilter* (electrical precipitator). On its collector plates with a total area of 25 m^2

the carry-over from the scrubber is deposited. The filter collector plates may be washed with water and detergent by a movable spray nozzle system. The bottom of the filter unit collects the wash solution containing the deposited activity, which will normally be discharged as waste.

An *absolute filter* (Gloved-box-filter "Luwa Patent Poelman") is connected to the exit of the electrofilter and provides an additional safety barrier. From here the off-gases pass a *gas meter* and are released to the outlet filter of the hot-cell ventilation. The off-gases are then diluted in the off-gas system of the reactor institute (10^6 m^3/h) and released through the 70 m stack.

6. — Experimental

6.1. — Preliminary Experiments.

A preliminary furnace consisted of an interchangeable grate of 3 mm diameter Nimonic-80 wire, placed on a steel support with a gas tube at the bottom. Placed on top of the support was an opaque quartz tube of 80 mm inner diameter surrounded by an 8 turn water cooled induction coil. A motor generator supplied the necessary medium-frequency power at 10 kc, the heat of combustion being removed partly by the cooling water of the induction coil, partly by radiation.

Graphite slugs of 60 mm dia., 300 mm long were burned in various mixtures of nitrogen and oxygen. The uncooled grate withstood the temperature of 1,000° C only for a few hours. The damaged grate was then replaced by a water cooled one

Fig. 6. — Graphite cone resulting from burning a graphite rod in the furnace.

in the form of a Nimonic-80 plate with concentric, circular slits and a silver soldered cooling spiral at its lower side. This type behaved quite well under the given conditions of combustion. A maximum temperature of about 1,400° C was reached. Only the support of the grate plate which was not water cooled, showed some minor damage. In an improved construction the support was also provided with a cooling coil.

The combustion zone was sharply marked on the graphite rod. The graphite burned quite rapidly at the lower end in a length of about 150 mm, so that a sharp cone was formed; this cone was more pointed the higher the combustion temperature and the higher the enrichment of the oxygen in the gas mixture. Typical data of some representative experiments of the combustion of graphite rods are given in Table 1 :

TABLE 1. — Results from burning graphite in the preliminary installation with 10 kc generator

Power input kW	Temp. °C	Composition of feed gas	Quantity of feed gas liter/min	Percentage of total oxygen consumed	Graphite burning rate g/h
4	930-70	air	11.5	94	70
3.8	930	air	33	57	120.5
3-3.5	950	36 % O_2	30	51	176
2-2.5	930	50 % O_2	30	45	216
0-1	950	80 % O_2	30	43	330
0-1	1,000	O_2	30	39	387
0-7	1,200-1,400	O_2	30	59.3	570

For all the experiments the initial high frequency power was 10 KW. Once the reaction started, the power was adjusted to the values given in table 1, which were necessary to maintain the combustion temperature at a constant level. Temperatures were measured pyrometrically through the quartz tube.

The results of these experiments show clearly that it is necessary to raise the combustion temperature above 1,000° C to reach sufficiently high burning rates. The use of pure oxygen seemed to be necessary to reach the high temperatures. The analysed off-gases showed the composition given in table 2.

TABLE 2. — Composition of the off-gases in the preliminary installation

O_2	52.6 %
CO	10.9 %
CO_2	35.5 %

6.2. — IMPROVED EXPERIMENTS.

Based on the preliminary investigations various improvements were made in the furnace part of the installation. The grate was completely modified to a hexagonal arrangement (fig. 5). This allowed loosely sintered agglomerates of ash to fall through the grate, whereas these pieces plugged the preliminary grates. The elaborate cooling system of the new grate is also shown in fig. 5.

The quartz tube was replaced by a ceramic tube of magnesium oxide and the whole arrangement was contained in a water cooled stainless steel tube. This tube was equipped with two holes to allow air to enter the system and to sweep out the space between stainless steel tube and ceramic furnace tube (fig. 4). This air also helped to burn CO to CO_2 in the upper part of the furnace. The medium frequency motor generator was exchanged for a high frequency tube generator (1,000 kc, 12KW) with high impedance terminals.

With these modifications the off-gases showed a much better composition in so far as no CO could be detected, thus decreasing the possibilities of explosion hazards very much. The composition of the off-gases of several runs is given in table 3.

TABLE 3. — Composition of the off-gases in the final installation

Compound	Composition %						
	Run 1	Run 2	Run 3	Run 4	Run 5	Run 6	Mean
$O_2 + N_2$	73	73	70	72	80	84	$O_2 + N_2$ 80.5
CO_2	24.5	27	29.5	25	20	15	
	Run 7	Run 8	Run 9	Run 10	Run 11	Run 12	
$O_2 + N_2$	90	90.5	81.5	86.0	79	86.5	CO_2 19.0
CO_2	9.7	9.9	18.5	14.7	21	13.5	

No CO was detected in these experiments.

If pure graphite rods were burned, an ash residue of approximately 0.5 per cent of the weight of the graphite was obtained. If Dragon fuel containing uranium and thorium carbides was burned, the following typical composition of the ash was obtained (table 4).

In a single run 92 % of the uranium and thorium were found in the ash. In a series of succeeding runs a yield of fuel ash close to 100 % was obtained, since the ash hold-up of the furnace is only a few grams. The carry over of light ashes (containing uranium) was about 0.2 % and was dissolved in the bicarbonate-scrubber by complexing action.

TABLE 4. — Composition of ashes from "Dragon-type" fuel

Compound	Composition in %				
	Run 1	Run 2	Run 3	Run 4	Mean
C	5.8	4.6	0.5	3.9	3.9
U_3O_8	16.3	16.6	16.9	15.7	16.4
ThO_2	77.4	77.6	82.0	78.7	78.9
Fe_2O_3	0.7	0.6	0.7	1.4	0.85

Experiments were made with the fuel ash in order to find an appropriate dissolution method. The methods tried and the results obtained are given in table 5.

We believe that a solution of concentrated HNO_3 — 0.1 *N* NaF — 0.2 *N* $Al(NO_3)_3$ would be most suitable to dissolve the uranium and thorium oxides and to separate them from the graphite residue.

TABLE 5. — Dissolution of fuel ash

Reagent	Dissolution of fuel component		
	Uranium	Thorium	Graphite
HNO_3	Totally	Partly	None
Oxidation at 1,100° C with O_2, fusion $KHSO_3$ or $K_2S_2O_7$	Quantitative	Quantitative	Quantitative
HNO_3-NaF-$Al(NO_3)_3$	Quantitative	Quantitative	None
NaF + HNO_3	Quantitative	Almost quantitative	None
HNO_3-HF	Quantitative	Partly	None

In a run lasting 120 hours a mean burning rate of 770 g of fuel per hour was achieved, this using an oxygen flow of 34 liters per minute; combustion temperatures up to 1,800° C were measured. About 70 % of the administered oxygen was consumed in this experiment.

A tracer experiment with slightly irradiated uranium-thorium carbide fuel showed no activity in the off-gas leaving the scrubber. The activity in the scrubber, mainly due to iodine and ruthenium was about 0.1 % of the total activity being present.

Experiments with coated particle fuel showed, that pyrolytic carbon coated fuel was readily decomposed, while silicon carbide coated particles were hardly affected. However, in the latter case the method is very useful for concentrating a large amount of highly active material without laborious and expensive procedures.

7. — Present state of development

The furnace and off-gas cleaning system has been installed in a hot-cell and operated with inactive fuel being now ready for active work. Modifications on the furnace were made in order to permit rinsing ot the whole furnace with decontamination agents and to permit disassembly of the furnace with the aid of the hot-cell manipulators.

Burning experiments with highly irradiated "Dragon" fuel specimen from post irradiation experiments will be done shortly thus leading to final results pertinent to the off-gas treatment system.

Acknowledgment

The authors wish to thank Mr. M. Oppliger for his very helpful assistance.

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SOME ASPECTS OF VOLATILITY PROCESSES FOR REACTOR FUEL ELEMENTS CONTAINING CARBON (1)

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ABSTRACT

Experimental results related to processing of high temperature gas-cooled reactor fuels containing carbon are discussed. Various reagents, such as nitric acid, oxygen, carbon dioxide and steam have been used to treat this fuel type. It was also possible to halogenate directly uncoated uranium carbide by means of hydrofluoric or hydrochloric acids. The compounds so formed can be further converted to volatile halides and especially to hexafluorides achieving a high decontamination.

Of the processes investigated, the use of the nitric acid vapor opens a new way for both dry and aqueous reprocessing.

1. — INTRODUCTION

This paper summarizes data obtained during studies performed on the reprocessing of graphite or carbon-base fuel elements. These results have been obtained in the scope of a program sponsored by Euratom, on ceramic fuel processing by halide volatility.

The data and the observation gathered during these studies permit us to draw conclusions about the alternative methods to be used for different types of fuel.

Reprocessing by halogenation is based on the volatilization of the uranium and plutonium halides from non-volatile fission products, which remain as a solid residue. A distinction must be made between the process which leads to the formation of uranium hexafluoride and the process which forms uranium penta-hexachlorides. The former is being developed, technologically speaking, and industrial plants are planned to be constructed in the near future; the latter is still subject to laboratory studies. A higher decontamination is achieved by the fluoride volatility process, compared to the chloride process which presents some difficulties in separating the zirconium and niobium volatile chlorides from the uranium.

(1) Research performed under the contracts 008-60-4 and 049-61-4 sponsored by the Euratom/United States Joint Research and Development Program.

(2) The principal investigators having contributed to the realization of the works presented are : G. CAMOZZO (a), A. FRANCESCONI, R. HEREMANS, G. PIERINI (a), J. SCHMETS, Ph. SPEECKAERT, A. STIENEN (b), J. BROOTHAERTS, F. COENEN, R. HARNIE, C. LAMBIET, A. LEURS and J. VANDERSTEENE.

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(b) S. A. Belgo-Nucléaire delegate at C. E. N. / S. C. K., Mol.

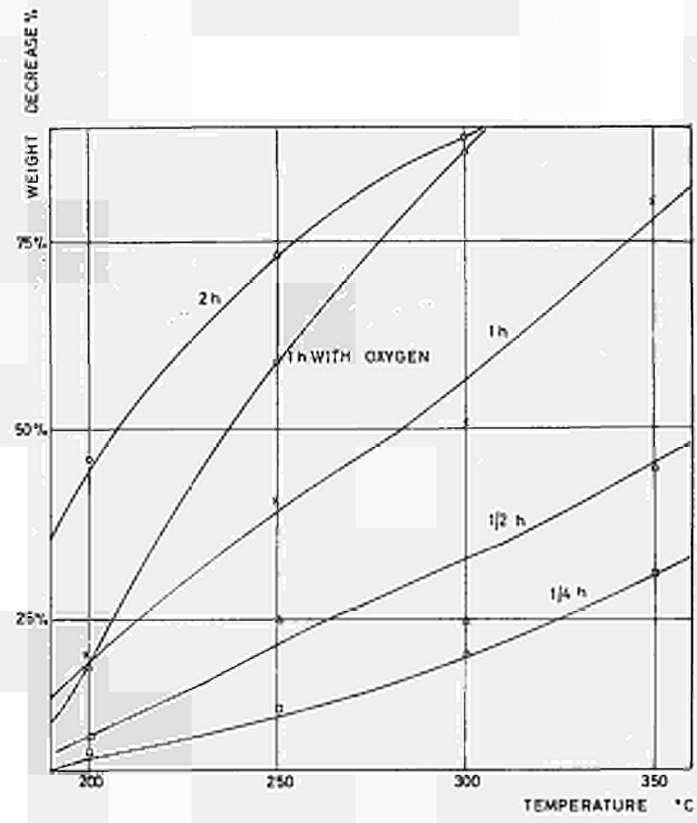

FIG. 1. — Oxidation of carbon by nitric acid vapors.

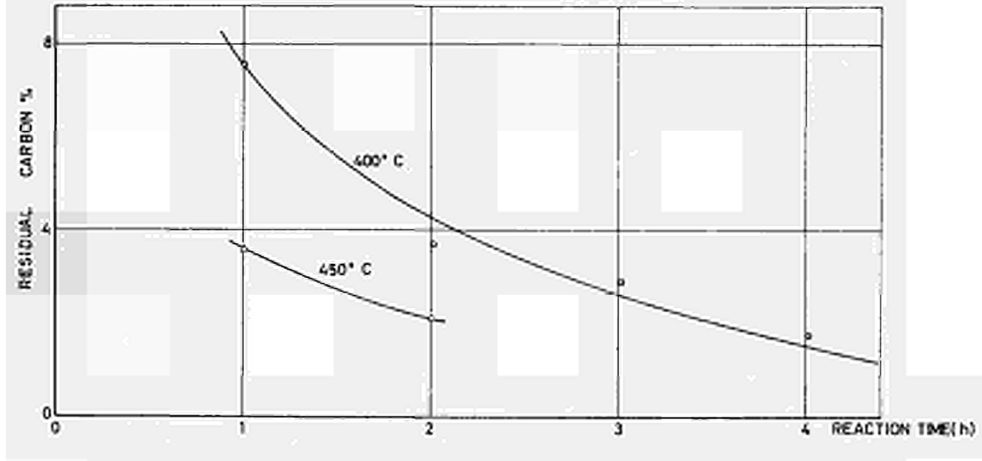

FIG. 2. — Oxidation of UC to UO_3 by means of nitric acid vapors and oxygen.

In many cases, a previous step of pulverization is needed in order to facilitate the halogenation and, particularly, to eliminate the carbon coating generally used for HTGR fuels.

Hereafter, aspects of preliminary treatment (a common step for both aqueous and non-aqueous reprocessing), fluorination, and chlorination of the fuel will be examined.

2. — Treatment of Fuel Containing Carbon Further Treatment of the Obtained Oxides

Different methods and reagents have been used in order to eliminate the carbon or to prepare the respective halides.

The reagents used in the course of our studies have been : nitric acid vapor, oxygen, carbon dioxide and steam.

2.1. — Fuel Treatment by Nitric Acid Vapors.

Nitric acid vapors permit a rapid elimination of the carbon which is associated with different fuels. Simultaneously, the uranium in the form of oxide or carbide is converted to UO_3 (the ε phase). The same reaction causes a destruction of the crystallographic structure. This step specially favours, in the case of mixed oxides, the subsequent conversion to fluorides.

2.1.1. — *Elimination of carbon.*

Fig. 1 represents the relationship between the percentage of the carbon eliminated by means of nitric acid vapor and the reaction temperatures. The curves give an idea of the reaction rate at different temperatures and for different reaction times. It is evident from these curves that the presence of oxygen in the nitric acid vapor favours appreciably the rate of reaction.

The elimination of the last traces of carbon present in uranium monocarbide appears feasible by slightly increasing the temperature or the reaction time. Fig. 2 shows how the residual carbon is reduced to 4 and 2 % of the original amount in UC when the temperature is raised to 400 and 450° C respectively.

2.1.2. — *The behavior of UO_2 and UC in the presence of nitric acid vapor.*

The dense compact UO_2 and UC are transformed into the ε phase of UO_3 by treatment with nitric acid vapor between 320 and 400° C. This reaction is favoured by oxygen, which probably prevents the dissociation of the nitric acid vapor.

Formation and stability conditions of ε UO_3 have been studied thermogravimetrically. The behavior of UO_2 when submitted to nitric acid vapor at increasing temperature is shown in Fig. 3. At the beginning of the reaction an increase in weight is associated with a heat release, due to the absorption of the vapor. The desorption causes the opposite phenomenon. At higher temperature there is a progressive

saturation in oxygen of the UO_2 lattice, followed by the formation of U_3O_8 and UO_3. Ultimately, above 400° C UO_3 decomposes.

The rates of reaction of UO_2 to UO_3 are given in Fig. 4. Also in this case, a significant increase in the rate is observed when oxygen is present. The reaction is similar with uranium carbide. As mentioned above, the elimination of the last traces of carbon, when required, will need perhaps a higher temperature of reaction.

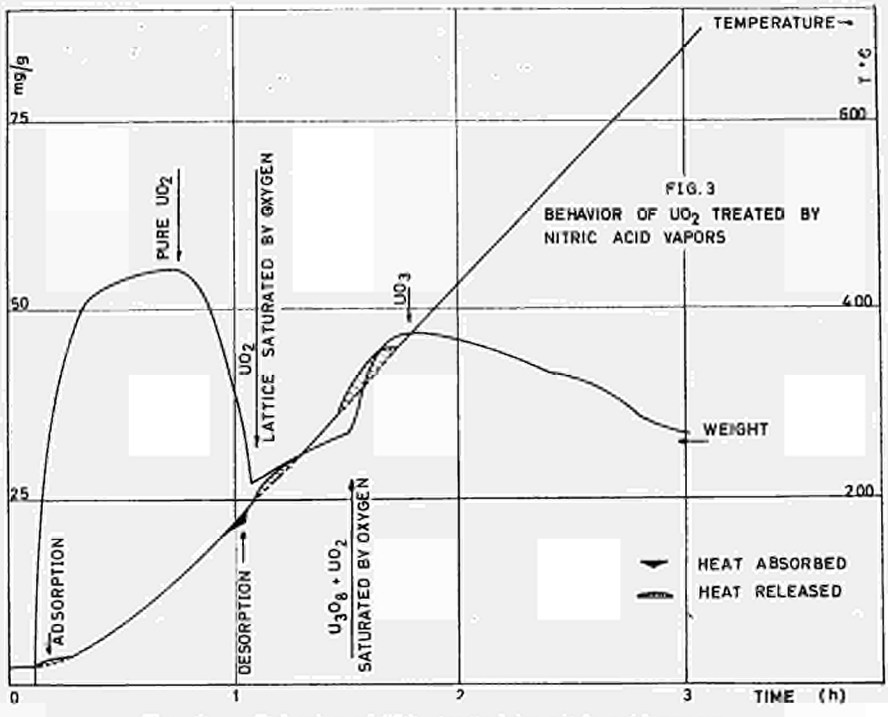

FIG. 3. — Behavior of UO_2 treated by nitric acid vapors.

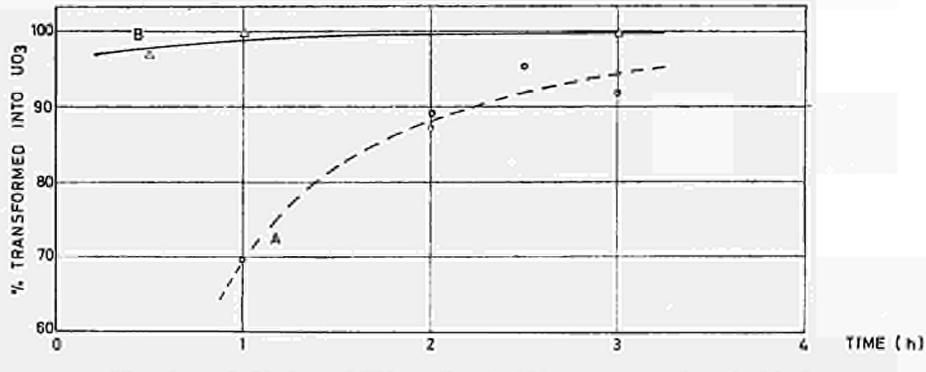

FIG. 4. — Oxidation of UO_2 pellets to UO_3 between 350 and 390° C.
A : HNO_3 vapors.
B : HNO_3 vapors associated with oxygen.

The thermal stability of ε UO_3 has also been investigated (Fig. 5). When ε UO_3 is calcined in air at 500° C, a stable phase $UO_{2.8}$ is formed. At higher temperature U_3O_8 is obtained.

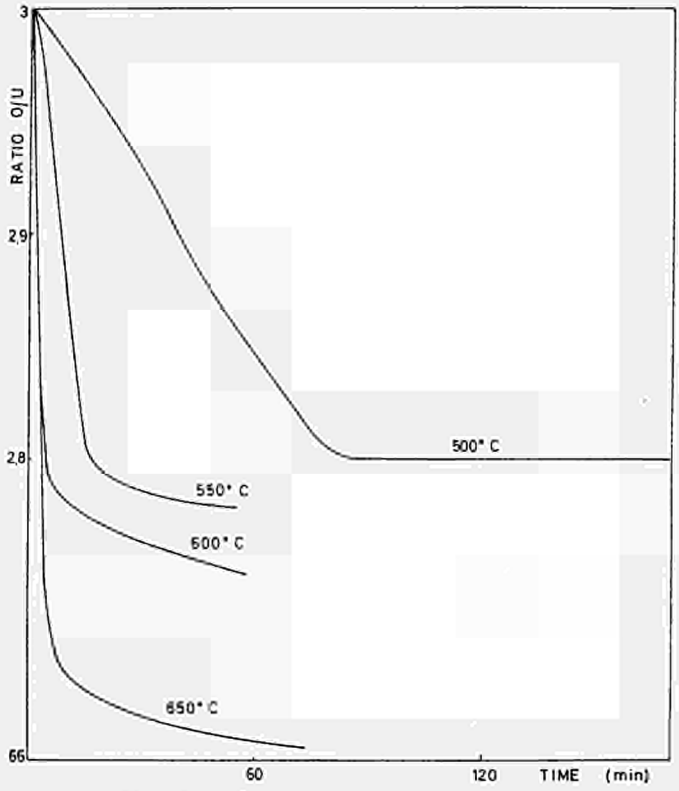

FIG. 5. — Decomposition of UO_3 in air.

2.1.3. — *Further treatment of the uranium trioxide.*

UO_3 can be submitted to different reagents in order to be transformed into fluorides. The following reaction has been studied

$$UO_3 \xrightarrow[300^0\,C]{SF_4} UF_6$$

When using SF_4, UF_6 can only be obtained from hexavalent uranium (UO_3 or UO_2F_2).

Experiments have shown that the rate of this reaction was rather slow; moreover, in the case of plutonium mixtures, it resulted in an uncomplete separation.

Other cycles can be used if UO_3 is used as starting product. UO_3 is thermally decomposed to U_3O_8, and the following reaction sequences can be applied :

$$- U_3O_8 \xrightarrow{HF} UF_4 . UO_2F_2 \xrightarrow{F_2} UF_6$$

$$- U_3O_8(PuO_2) \xrightarrow{HF} UF_4 . UO_2F_2(PuF_4) \xrightarrow{ClF_3} \nearrow UF_6(PuF_4) \xrightarrow{F_2} PuF_6$$

These reactions will be used in the α—γ pilot facility for spent reactor fuel processing at Mol.

2.1.4. — *Technological development of the process.*

As far as the oxidation to UO_3 of ceramic fuel is concerned, a technological development up to some hundred grams of fuel has been made. This reaction has mainly been studied in vertical reactors. Systematic experiments have shown that for pure UO_2, this process is very delicate because of the small range of temperature in which UO_3 is formed and doesn't decompose.

However, this conclusion is not to be applied either to the elimination of carbon or to the uranium-thorium oxide mixture. In this situation it is expected that the proposed method is of easier application.

In fact, during the further treatment of these oxides to fluorides, uranium is not required in form of trioxide, as it was necessary for the conversion to UF_6 by means of SF_4.

Moreover, the oxides obtained by the nitric acid vapors at a relative low temperature are much more amenable for leaching than those obtained by oxidation at very high temperature (800-1000° C) by means of oxygen. In others words, the chemical reactivity of the oxides formed by the nitric acid vapor is still high and permits easy treatment.

Corrosion problems associated with the use of nitric acid vapor have been investigated. Incoloy (Ni-onel), Corronel 230, titanium and tantalum are considered as useful materials.

2.2. — THE OXIDATION OF THE URANIUM CARBIDE BY AIR, STEAM AND CARBON DIOXIDE.

2.2.1. — *Basic study.*

Fig. 6 and 7 give an idea of the behavior of the reactions between UC and oxygen, air, carbon dioxide and water as deduced from thermogravimetric experiments at increasing temperatures.

The oxygen causes a fast and uncontrolled reaction. The reaction with air is slower and leaves a residue of carbon which is progressively eliminated above 500° C. These two reactions lead to U_3O_8, while the reactions with carbon dioxide and steam lead to UO_2.

It has been observed that the elimination of carbon starts at 650° C or at 500° C when respectively carbon dioxide or steam are used as oxidizing agents.

2.2.2. — *Development.*

The high reactivity of UC would suggest that its chemical transformation might constitute an easy technological operation. In fact, the high affinity of uranium monocarbide makes the initial surface reactions very fast. This gives rise to a sintered coating which becomes impermeable to the reagent, inhibits the pulverization, and promotes the formation of granular material.

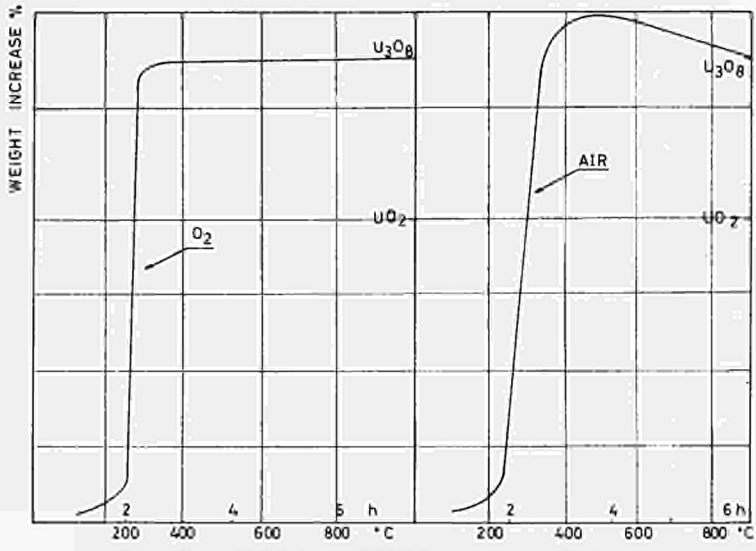

FIG. 6. — Oxidation of UC by oxygen and air.

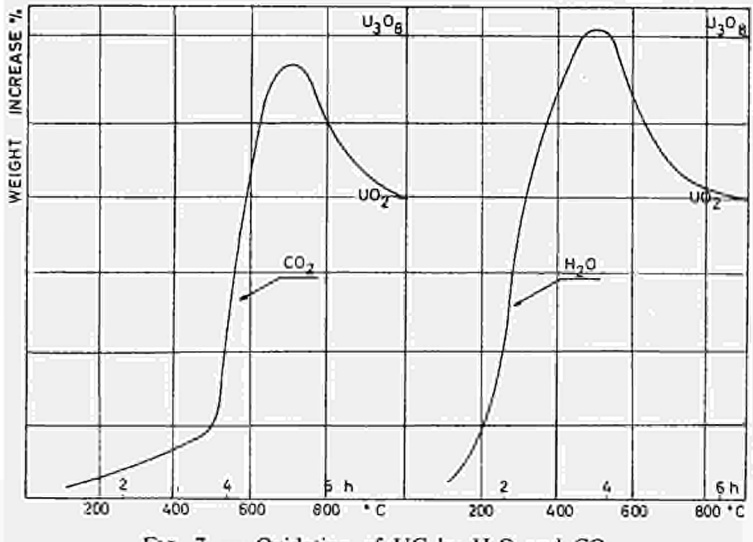

FIG. 7. — Oxidation of UC by H_2O and CO_2.

The different reactions described above have been investigated in a fluidized bed on high-density sintered uranium carbide. In all cases a superficial partial sintering has been observed as well as a certain impermeability to the gaseous reagents. Fluorination experiments on oxides obtained from carbides have not yet been performed.

Two observations can be made :

— The experience acquired during similar reactions on uranium oxides shows that fluorination of heterogeneous mixtures of powder and granules is possible;

— Conclusions cannot be drawn from the experiments before working on irradiated fuels.

In fact, during irradiation these fuels undergo a modification in their structure and in their composition. Results obtained by cross-checking indicate that irradiated fuels will be less sensitive to exothermic effects and that reactions will behave more smoothly.

3. — Hydrofluorination and Hydrochlorination of Uranium Carbide

3.1. — Hydrofluorination of Uranium Monocarbide.

3.1.1. — *Basic study.*

This reaction constitutes the first step of the following cycle :

$$UC \xrightarrow{HF} UF_4 \xrightarrow{ClF_3 \text{ or } F_2} UF_6$$

It appears, a priori, convenient for uncoated uranium carbide reprocessing. It is a complex reaction which produces hydrocarbons and hydrogen according to the schemes :

$$UC + 4HF \longrightarrow UF_4 + C + 2H_2$$

$$UC + 4HF \longrightarrow UF_4 + \frac{C_nH_{2n+2}}{n} + \frac{n-1}{n} H_2$$

From a thermodynamic point of view, the first of these reactions is the most probable at high temperature. It was in fact experimentally confirmed. On the other hand, the reaction giving hydrocarbons causes the formation of heavy paraffins. These can, at a certain temperature, inhibit the reaction by making a coating on the particles. This effect is predominant below 200° C as shown in Fig. 8.

Similar experiments have been carried out with the aid of liquid hydrofluoric acid. The reaction proceeds according to the above mechanism.

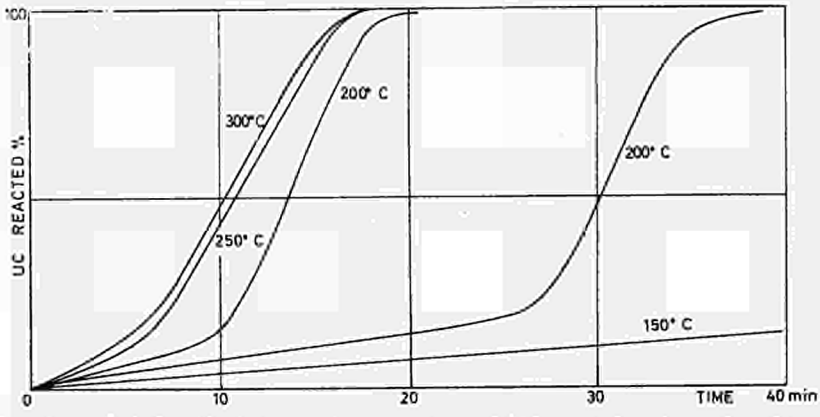

FIG. 8. — Relationship between the percentage of UC reacted and reaction time.

3.1.2. — *Development.*

Two series of experiments in fluidized bed have been effected : some on low density carbide, others on high density carbide. With low density carbide, the reaction occurs normally; high density carbide gives a formation of caking by a superficial partial sintering. Experiments are carried out according to techniques utilized in similar cases with oxides. Remarks which have been made for carbide oxidation can also be applied here.

3.2. — URANIUM CARBIDE HYDROCHLORINATION.

3.2.1. — *Basic study.*

Uranium monocarbide has been hydrochlorinated by means of dry and wet hydrochloric acid. A sublimation of uranium has been observed. Moreover, a fraction of uranium is retained by the residual carbon. It seems preferable, for the chlorination reprocessing either to make a preliminary oxidation, or to chlorinate the uranium directly to higher valences according to the following schemes :

$$UC \xrightarrow{ox.} UO_x \xrightarrow[Cl_2]{CCl_4} UCl_{5-6}$$

$$UC \xrightarrow[Cl_2]{CCl_4} UCl_{5-6}$$

4. — CONCLUSIONS

Amongst the proposed procedures of chemical attack for carbon-base fuel elements, the action of the nitric acid vapors opens a new way for both dry and aqueous reprocessing.

The utilization of the halogenation procedures offers various possibilities which are to be examined and checked for every type of fuel.

As far as the high-temperature gas-cooled reactor fuels are concerned, the elements differ in their micro- and macrographic structure and in their composition and therefore each type of fuel element will require the development of a specific technology.

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PREPARATION OF URANIUM DIOXIDE AND CARBIDE PARTICLES BY SOL-GEL METHODS

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ABSTRACT

The process for the preparation of spherical uranium carbide or oxide particles consists of the following phases :

— preparation of a colloidal uranium (IV) solution, containing carbon powder whenever it is desired to obtain carbides;

— dispersion of the colloidal solution, under the form of droplets, in an organic solvent immiscible in water and gelation into spherical particles having the desired diameter;

— drying and thermal treatment of the gel particles.

During the last Geneva Conference a report was presented on the results of research carried out in the CNEN laboratories with a view to developing a sol-gel method for the preparation of spherical uranium and uranium-thorium oxide particles [1].

Such research was continued over the past months and the procedure used was extended also to the preparation of uranium carbides and mixed thorium-uranium carbides; moreover, single stages of the procedure were developed for the purpose of building a pilot plant with a capacity of a few kilograms per day of sphere-shaped uranium oxide or carbide particles and/or uranium-thorium particles (fig. 1). Research on thorium-uranium carbides is being carried out on account of Dragon Project and the results obtained appear in another paper of this symposium.

Within the framework of researches being conducted on account of Dragon Project, a small plant was assembled for the continuous extraction of nitric acid, with regeneration of the anionic liquid exchanger used in the preparation of acid-deficient thorium nitrate and uranyl nitrate solutions. The plant has been operated for several days without any special difficulties arising.

The acid-deficient uranyl nitrate solution is then evaporated at temperatures not exceeding 80° C up to concentrations of 3-3.5 moles of uranium per litre and is subsequently reduced with hydrogen. Such reduction may be carried out either in a discontinuous autoclave or in a continuously operating one, by placing the solution to be reduced in contact with hydrogen under pressure, with the help of a catalyst consisting of platinum on alumina powder or grains [2]. It was not found necessary to add stabilizing substances to the uranyl nitrate solution. In batch reductions — and operating at room temperatures, at a pressure of 30 Kg/cm^2 — a reduction yield of more than 98 % is attained in approximately 60 minutes. It should be borne in mind that the period of time required for reduction is greatly affected

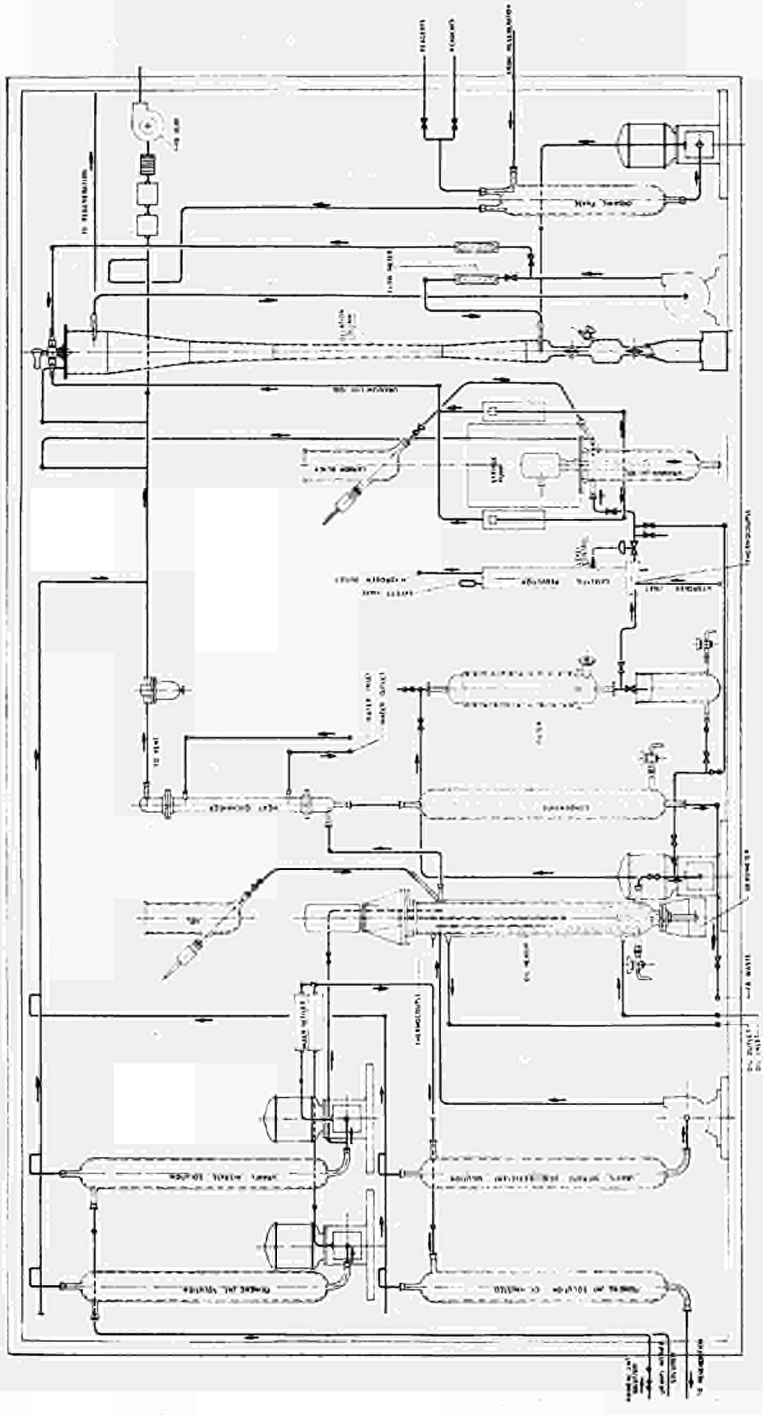


FIG. 1. — Equipment flowsheet for the preparation of uranium dioxide particles by Sol-Gel method.

by the contact between aqueous solution and catalyst and, consequently, by the efficacy of agitation. The colloidal uranium (IV) solution is formed during the reduction phase. Although the phenomena taking place in the course of reduction have not been studied in detail as yet, it was observed that long periods of contact lead to non-stable solutions or directly to the formation of gels. The viscosity of the colloidal uranium (IV) solution is inferior to that of the corresponding acid-deficient solution : such reduction in viscosity is particularly evident in the case of colloidal U-Th solutions. The colloidal uranium solutions do not present settling phenomena and do not oxidize : if maintained out of air contact they remain unaltered for many months. Finally, the characteristics of the colloidal uranium (IV) solution depend to a very large extent on the uranium (VI) content; if such content exceeds 5 % the subsequent gelation of particles proves quite difficult to achieve and the consolidated particles do not, as a rule, withstand thermal treatments. The continuous reduction of concentrated, acid-deficient uranyl nitrate solutions has not been developed up to now but no particular difficulties are expected at this stage of the process.

For the preparation of uranium carbide particles, carbon-powder must be added to the colloidal uranium (IV) solution.

Following the advice of Dragon Project experts, we are making use of carbon-black "United 15", which does not require special techniques for incorporation into the colloidal solution at room temperature. Among the incorporation methods practicable in laboratory experiments, we adopted the one involving a porcelain ball mill. It is thus possible to obtain a perfect distribution of the carbon, with fragmentation of the aggregates. When preparing the particles on a larger scale, the ball mill will be replaced by a mechanical disintegrator — of the type used in the pigment industry.

1. — GELATION

In sol-gel processes the formation of sol-gel particles of ceramic materials is attained by dispersing the sol inside a substance immiscible in water, thus causing the consolidation or gelation of the sol droplets. This gelation process is generally carried out by raising the pH or by extracting water from the sol. The raising of pH can be arrived at by neutralizing the nitric acid existing in the sol droplets (with ammonia, for instance) or by extracting the nitric acid itself. Gelation of the colloidal uranium (IV) solution droplets is performed continuously by using a facility with a cone-shaped tower, similar to that already studied at the Oak Ridge National Laboratories [3]. The extraction of water has the advantage of causing a drastic decrease in the particles' diameter during the gelation phase and, therefore, when using the above-mentioned facility it facilitates considerably continuous operation.

However, the particles prepared in this manner contain all the nitric acid already present in the colloidal uranium (IV) solution; the subsequent drying and calcinating treatments thus become particularly critical, in that the decomposition of the nitric

acid provokes the oxidation of the uranium (IV) and the breaking of the particles. On the other hand, it is impossible to proceed with the washing of the colloidal particles, as for instance using a diluted ammonium hydroxide solution, since a re-hydration of the particles would quickly cause them to break.

Gelation through neutralization of the nitric acid present in the colloidal uranium (IV) solution with ammonia could be performed from the outside, by adding ammonia to the organic phase, or from the inside, similarly to what is being done in the process for the preparation of Th-U oxide particles, where a substance is added to the colloidal solution which, in the long run — (or owing to an increase in temperature)— generates a certain amount of ammonia as a decomposition product [4].

It is evident that gelation from the inside would yield the best results. It should be borne in mind, however, that the quantity of nitric acid existing in the colloidal uranium (IV) solution is 3 to 4 times that normally contained in the colloidal Th-U solutions used in the above-mentioned process. Thus, in order to achieve gelation of the droplets within a reasonable period of time, it would be necessary to add a substantial amount of gelation agent, with the consequent partial gelation of the sol mass, prior to its subdivision into droplets.

It was therefore deemed useful to cause gelation by extracting the nitric acid from the sol droplets, adding an anionic liquid exchanger to the organic phase. If the extraction of the nitric acid is done slowly enough, so as to avoid concentration gradients between the external and internal layers of the sol droplets, it is easy to obtain spherical uranium oxide gel particles without hollows which do not raise any particular difficulties during the subsequent thermal treatments (fig.2). In fact, a thermogravimetric analysis of the uranium (IV) gel particles has shown that there is a uniform weight loss from 120° C up to almost 700° C.

So far gelation has been carried out in batches and no special difficulties were encountered, even if the colloidal uranium (IV) solution contains the amount of carbon necessary to prepare uranium carbide.

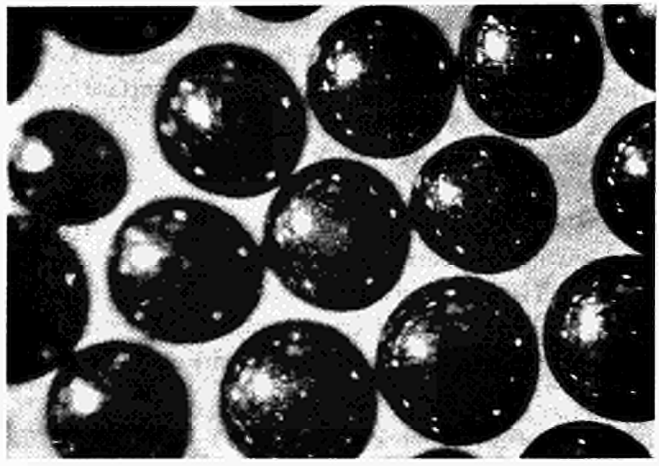
The organic phase used was Alphanol 79, a Shell solvent consisting of a high molecular weight alcohol mixture, saturated with water at room temperature and containing Primene as liquid anionic exchanger and SPAN 85 as tensioactive agent; in our specific experimental conditions the period of time required for gelation is approximately 15 minutes.

Regeneration of the organic phase has not yet been studied in detail : we think that it may be carried out continuously in a mixer-settler with an aqueous sodium hydroxide solution.

2. — Drying and thermal treatment

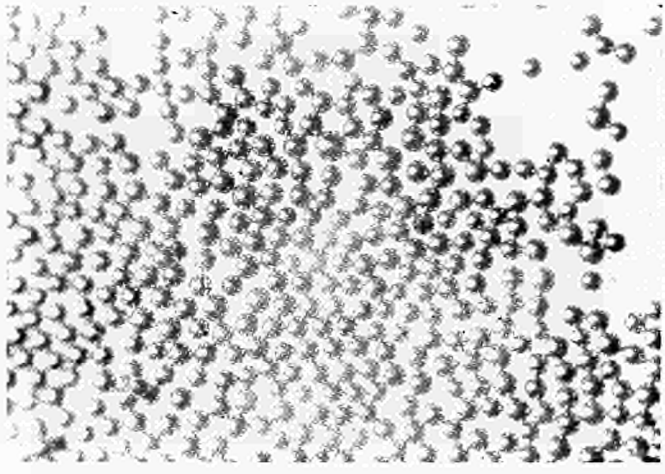
The spherical uranium (IV) gel particles are dried and fired in hydrogen inside an electric-resistance furnace, at a programmed temperature.

Temperature is increased up to 300° C at a rate of 0.5° C/min. and up to 1,000° C at 3° C/min; finally, it is maintained constant at 1,000° C for one hour.



40 ×

FIG. 2. — Spherical particles of UO_2 as gelled.



12 ×

FIG. 2. — Spherical particles of UO_2 1 h, 1,000° C in hydrogen.

When preparing uranium carbon particles, the carbon-bearing gel particles are dried up to 700° C in hydrogen, making use of the same furnace and heating velocity as that mentioned above. The dried particles are then transferred into a high temperature furnace, where they are submitted to 1,700° C under vacuum. Since we did not have at our disposal an adequate furnace, this latter thermal treatment was performed at temperatures not exceeding 1,300 °C and the carburization reaction could therefore not be completed. It should be kept in mind that, in order to dry the uranium (IV) gel particles, a temperature as high as 700° C is required, so as to avoid reoxidation of the uranium during the transport of the particles from the drying oven to the carburization furnace.

The spherical uranium dioxide particles produced by the above-mentioned sol-gel process lack porosity and have a density (determined in CCl_4) of 98 % with respect to the theoretical density (fig. 3). Their specific surface area (BET) is less than 1 m^2/g.

An X-ray diffraction analysis has shown that the dioxide crystals have a diameter of 5,000 Å.

Particles up to 500 microns in diameter were produced. As already mentioned, the gelation phase has not yet been completely developed, in particular as regards the sol subdivision which is still being done by means of a rather rudimentary device. It was therefore impossible to perform any granulometric analysis in order to control the homogeneity of the diameter in the produced particles.

3. — Conclusions

The procedure described above makes possible the preparation of spherical uranium dioxide or uranium carbide particles from aqueous uranyl nitrate solutions containing any amount of free acidity. This method was extended, on account of Dragon Project, also to the preparation of spherical thorium-uranium carbide particles.

The application of this process also to the preparation of spherical uranium-plutonium particles with rather high plutonium concentrations might give rise to some difficulties, owing to the reaction between uranium (IV) and plutonium (IV) at the time when plutonium is added to the colloidal uranium (IV) solution. Some preliminary tests were performed, simulating plutonium with cerium and we prepared, with satisfactory results, spherical uranium-cerium particles (at 3 % at of cerium).

It will be possible to evaluate the sol-gel method only after having started to run the pilot plant, which we hope to build and begin operating within one year.

Acknowledgment

The authors wish to express their appreciation and gratitude to Ing. M. Fiorelli of Eurex for having designed the pilot plant.

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BASIC STUDIES RELATED TO THE DEVELOPMENT OF THE SOL-GEL PROCESS

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ABSTRACT

A review of the development of the KEMA Sol-Gel process is given.

The external gelation method, in which ammonia is added from the exterior to a sol droplet has been successfully used for 10 μm particles. It fails, however for particles of larger size, viz. 250-1,000 μm.

The use of an ammonia donor, e.g. hexamethylene-tetramine, dissolved in the sol, leads to a successful preparation of particles of the larger size. A washing step, in which nitrate is removed, must be included in the procedure.

Both oxide particles and oxide-carbon mixtures, ready for reaction and heat treatment can be prepared.

A few data characterising hydroxide precipitates, sols and oxide particles are given.

Photographs illustrate the influence of the composition of the washing liquid on the formation of defects in oxide particles.

1. — INTRODUCTION

A sol-gel process for the preparation of spherical UO_2-ThO_2 particles of 5 μm diameter was developed at KEMA early 1959 and has been in use ever since this date for the production of reactor fuel with fully enriched uranium and material with natural uranium for technological experiments.

This process has been described [1], but the principles of it will be repeated here, since they formed the basis for further developments to be described.

A sol is prepared from freshly precipitated, well washed thorium hydroxide by peptization with a mixture of nitric acid and uranyl nitrate. This peptization takes place at the boiling point and is finished after about five minutes. The sol is concentrated by evaporation, to about 2.5 mol/l.

The sol is dispersed in the form of droplets by suitable agitation with an organic liquid, chosen for its density, containing a surface active agent to produce a stable emulsion of the water-in-oil type.

The droplets are solidified by interaction with ammonia. This ammonia is dissolved in the same liquid as used for dispersion. The emulsion is mixed with the ammonia bearing liquid and gelation takes place.

A dehydration step is necessary to strengthen the particles formed. It is carried out by distillation of water with carbon tetrachloride. After removal of the water the carbon tetrachloride is drained off and the particles washed with ammoniacal methanol.

There has been an increasing interest in spherical particles of larger size that might be used in dispersion elements or, after coating with carbon, as fuel for high temperature reactors.

In the latter case carbides or, eventually, oxides, will be used.

This interest has led to a modest effort in developing the process described above for the production of larger particles. This development was greatly aided by a contract from the Dragon Project.

The work carried out aimed at fulfilling the objectives by a natural development of the existing experience. Duplication of the work of others, notably the large scale efforts of the Oak Ridge National Laboratory, has been avoided as far as possible.

A process has been found, that can meet, after suitable development from the technological point of view, all requirements known at present.

It is, however, not offered here as the ultimate sol-gel process. Depending upon conditions of production, a combination of the several processes now under development will provide the most economic solution.

2. — Further development of the ammonia process

The first modification of the process is concerned with the dispersion step.

The necessary changes in stirring conditions when larger particles are to be produced, are not easily found. Nevertheless it has been shown that oxide particles of diameters up to 25 μm can be produced with but slight changes in the conditions.

For larger particles the droplets have been formed by means of dropping from a capillary, or stirring was applied where it was thought to be advantageous to have a broad spectrum of particle sizes present.

For the preparation of oxides no change in the sol preparation step is necessary but there are several possibilities for the introduction of carbon. In the absence of suitable mixing apparatus two ways of introduction of carbon have been used.

In the first one the carbon is dispersed in the thorium nitrate solution and entrained with the thorium hydroxide precipitate when this is produced. The mixed precipitate is taken through the usual peptization step. Good dispersion of the carbon is achieved. The filtration and washing of the thorium hydroxide, the most tedious step of the process, is made more difficult by the presence of carbon.

This difficulty disappears when the carbon is dispersed in the nitric acid-uranyl-nitrate solution used for peptization. This suspension can be used for the peptization in the same way as the solution.

Where suitable mixing equipment is available there are advantages (as found by Dragon Project workers) in dispersing the carbon in the sol that is ready for gelation. In this way the preparation of the sol and especially its concentration by evaporation after the peptization step are better controlled and the concentration can be carried somewhat further.

The peptization step, whatever method is used, is not easily described, since it is not exactly known what are the controlling variables. The skill and experience of the operator are important factors.

An attempt to describe this procedure is therefore not made.

The amount of uranylnitrate used for peptization is determined by the desired ratio of U to Th, nitric acid is added to obtain a nitrate content of 30-40 mole % based on total metal.

Nitrate and nitric acid are only used because uranyl nitrate is the normal starting material. Chloride and hydrochloric acid could be used as well if uranyl chloride were readily available and corrosion problems did not play a role. Indeed, in that case a major complication of the process would disappear, viz. the removal of nitrate.

In the ammonia process this removal is achieved during the washing with ammoniacal methanol. In larger spheres the removal of nitrate is insufficient and the nitrate ion, or ammonium nitrate, causes an explosivelike fragmentation of the spheres on heating at temperatures between 100 and 200° C. A nitrate removal step has to be introduced therefore, giving rise to its own special difficulties.

When it is tried to gel with ammonia, more and more failures are met with, when the diameter of the droplets increases. The failures appear for oxide spheres from 25 μm upwards and for oxide-carbon samples from 100 μm upwards and are found in all of the three stages : gelation, washing and drying. Quite a number of phenomena occur : rupture of a membrane of gelled material, showing the inner part of the drop to be still liquid, cracks of all kinds, deformation of droplets due to too slow gelation, hollow particles.

However the particles never fall apart as a powder. The impression is formed that several types of stress distribution are produced in the gelled particles. Once these stresses are relieved by formation of the cracks belonging to the particular type of stress distribution, the remaining particles or particle fragments are stable.

It will not be attempted to describe in detail the various observations. A similar example is considered later on.

The occurrence of stresses points to phenomena of contraction or expansion in the gel, unequal in magnitude within the sphere. Most probably these are caused by inequalities in amount of gelation and concentration differences within the sphere and between the sphere and the surrounding medium.

It is not difficult to see how these arise. It is tried to scale up the particle radius from 5 to 500 μm, hence a factor of 100. The mass has therefore increased by a factor 10^6, requiring 10^6 times more ammonia per sphere. The surface of the drop has increased by the factor 10^4 and to gel the particles in the same time the ammonia flow to the interior of the drop has to be increased a hundred fold, requiring a hundred times higher outside concentration. Or, if the same concentration difference was applied, the gelation would take a hundred times longer.

Another difficulty arises. On first contact of the drop with ammonia a gel membrane is formed around the drop. This not only hinders the entrance of ammonia but allows an osmotic inbalance, giving rise to a water flow either to the inside or

to the outside of the particle. A flow of water to the inside causes the rupture of the membrane, a flow of water to the outside can lead to a wrinkling of the surface. Both phenomena have been observed.

In case the water is driven out of the particle, it makes a difference whether a surface active agent is present in the outer, organic liquid. If it is present, the water is soon dispersed as an emulsion.

If it is not present, a drop of water remains attached to the gel drop and increases in size. It will preferentially dissolve ammonia from the organic phase, thus introducing further inbalance in the system.

If the gelation has proceeded satisfactorily, similar phenomena can occur during the washing step. It has been shown by chemical analysis that practically all of the nitrate is removed from the particle, showing it to be quite porous.

Particles that have survived both stages may still crack during drying or, having withstood the drying at temperatures between 100 and 200° C, may crack on reexposure to room conditions.

It was felt from these observations that a process worked out to overcome these difficulties would not only require a long time for development but, when successfully developed, would require delicate control and long times.

The considerations formulated above led to the orientation of the work into another direction.

3. — The method of internal gelation

The difficulties arising from concentration gradients could be overcome if it were possible to supply ammonia at equal rate to all parts of the sol droplet.

This problem has been solved for the production of precipitates in analytical chemistry by the method known as precipitation from homogeneous solution [2]. This method has been used also for the preparation of UO_2 particles [3].

The same procedure was tried for the present problem. An ammonia donor can be incorporated in the gel, without causing immediate precipitation, and, when ammonia is released from the donor, gelation occurs at the same time throughout the whole of the particle. We have termed this "internal gelation" by contrast with the procedure in which ammonia diffuses into the droplet from the outside.

The well known ammonia donors hexamethylene-tetramine, urea, acetamide and potassium cyanate can be easily dissolved to concentrated solutions that can be satisfactorily mixed with the sols, both with and without carbon. Acetamide and potassium cyanate have proved to be less suitable for gelation under the present conditions. The hydrolysis of urea proceeds with sufficient rate only at higher temperature, 95° C and higher. Hexamethylene-tetramine can be used both at room tempetature and at higher temperatures. Most of the experience has been obtained with this substance.

The concentration limits for the process are easily defined. The concentration of the solution of the ammonia donor, which is to be mixed into the sol, should

be high to avoid dilution of the sol but not so high that immediate gelation occurs, due to the high p_H of the solution. The ammonia donor should be in rather large excess with respect to the nitrate of the sol, because the hydrolysis is never complete. The rate of gelation at any temperature can be varied by adjusting the excess of the donor.

The dispersion of the sol in the organic liquid is not affected and, after suitable washing, the removal of the excess reagent by heating does not present any difficulties.

The choice of the organic dispersion medium is limited. It has been found that, with the sols studied, a too strong dehydrating action of the organic liquid on the sol has to be avoided. This difference with the experience at ORNL is not understood.

The lower alcohols, such as propanol and butanol are examples. As normally supplied, their dehydrating action is too strong and good particles are not obtained. When saturated with water good spheres can be produced but their surface remains weak because there the material tends to be peptized in the alcohol that contains much water.

The alcohol 2-ethyl-hexanol, extensively used at Oak Ridge National Laboratory in a process that seems to be pure dehydration, is a suitable compromise between the properties of drying action and water solubility.

Two types of procedure can be envisaged when the method of internal gelation is used.

In the high temperature variant, cold sol, with gelling agent, is dropped into the heated organic liquid. The hydrolysis proceeds at a rapid rate at high temperature and good particles are formed within a minute. The dispersion liquid used so far has been tetrachloroethylene, chosen for high boiling point, high density, low solubility of water and non-inflammability. This substance is poisonous and work should be carried out in a fume cupboard. Early experiments with xylene were not successful but the possibility of using this and comparable liquids e.g. mineral oils, is certainly not excluded.

Gelation can be carried out at 100° C but this temperature should not be exceeded too far, because the evaporation of water from the sol (or gel) droplets will cause much the same defects as experienced on dehydration.

To ensure good dispersion a surface active agent is present in the organic phase.

Particles produced according to this process have been treated successfully by Dragon Project workers and at CEN, Mol, Belgium. This part of the process is not discussed here.

Urea can be used instead of hexamethylene-tetramine in this variant.

The low temperature variant is commonly called the Winfrith variant by us because this modification was developed by Dragon Project workers, to which may be referred for details. Their experience has been fully confirmed here.

In this type of process use is made of the slow hydrolysis of hexamethylene-tetramine at slightly elevated temperatures. This means that, to ensure good gela-

tion, a relatively large excess of the ammonia donor must be used and a somewhat longer time of residence applied, these conditions being, of course, related to each other. In fact, it is necessary to cool the sol-donor mixture to prevent gelation during the time in which the sol drops are formed. Even then the viscosity of the sol changes markedly and only a relatively small amount of sol can be gelled continuously. As a dispersion liquid 2-ethylhexanol has been used. The lower density of this liquid and the longer time of residence makes it necessary to keep the particles suspended in an upward flow of the alcohol.

To prevent dehydration the alcohol is saturated with water. The impression has been formed that the best particles are produced at a temperature slightly above that at which the alcohol is just saturated with the water present.

As a first step a coating of gel might be formed around the drop, that prevents deformation during the subsequent gelation by ammonia production. (Such a first step occurs in the high temperature variant because the outer layer of the droplet gels at once on contact with the hot liquid).

The gelled particle should not come into contact with the dispersion medium at a lower temperature. The organic liquid then contains an excess of water, present as an emulsion which is taken up and tends to soften the outer part of the particles.

After gelation the particles are washed to remove the alcohol, nitrate is removed in an ammonia bath and the particles are ready for drying and heat treatment. Satisfactory oxide and carbide materials have been produced.

Finally, it should be noted that a recent patent search revealed that the principle of internal gelation has been proposed much earlier for the production of aluminium oxide catalyst spheres [4], although the explanation given differs somewhat from that given here while the process conditions are different also.

4. — Some properties of precipitates and sols

In the following paragraphs some data regarding fundamental properties of sols and gels will be given. No systematic study has as yet been made and what is given here only forms the results of a few scouting studies.

It has already been noted that the sol preparation step is not well understood, this leads to a certain irreproducibility in the gelation properties. Consequently the exact amount of ammonia donor to be added has to be found by experiment for every new batch of sol. It is felt that a deeper understanding of these steps might remove these difficulties.

A study of the behaviour on sintering may aid in defining optimal heat treatments. This would seem to be useful especially for oxide material. The reaction step in the preparation of carbide may lead to the disappearance of relationships between the starting material and the product.

Starting material for the present procedure is thorium hydroxide, precipitated from nitrate with ammonia. This procedure can be carried out with many variants,

which have not been fully investigated. As might be expected, it is best to keep temperatures low and use the precipitate in the freshly washed condition.

It is remarkable that the specific surface of the precipitate is very low after a moderate heating, while at the same time the crystallite size is low. In this case the precipitate forms a glass like material, that resists peptization.

Table 1 gives some results. In case A thorium nitrate has been added to ammonia, in case B the reverse procedure was applied. The temperatures are given at which the precipitate was heated for 3-3½ hrs.

The specific surface S, expressed in m^2/g was determined from low temperature nitrogen adsorption (1), the crystallite size from X-ray line broadening (2).

TABLE 1. — Properties of heated precipitate

A			B		
Temp.	S	*d*	Temp.	S	*d* (Å)
70	16	15	80	0.5	15
190	2.4	15	200	0.5	20
300	0.9	30	300	0.5	30

The crystallite size of precipitates left at the atmosphere at room temperature is about 20 Å.

The sols of thorium oxide are characterized by high concentration, low viscosity and insensitiveness to the addition of electrolyte.

The viscosity depends strongly on p_H, only a slight increase in p_H leads to a gradual thickening of the sol, ending with the formation of a stiff gel.

From X-ray investigations it would seem that the major fraction (if not all) of the oxide is present in the form of small cubes of 20-30 Å (3). The occurence of these particles would explain the low viscosity at low p_H. Other thorium species would seem to be present in only minor amounts, although it is not excluded that they might, even at low concentration, play a role in the gelation.

The process of peptization might be envisaged as one of leaching amorphous parts of the precipitate, whereby the crystallites are set free, at the same time acquiring a positive surface charge. When this process takes place at the boiling point, it may be accompanied by some crystal growth.

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(1) Determinations of specific surface area were carried out by Mr. G. J. ZONDERVAN.

(2) All determinations of crystallite dimensions of solids by ir. T. MARKESTEIN.

(3) The authors are indebted to prof. dr. J. A. PRINS, Technological University, Delft, and ir. T. MARKESTEIN for these determinations.

The sols can be evaporated to highly concentrated sols with still a relatively low viscosity. Under some conditions a phase separation occurs, whereby two immiscible sols are produced, differing in concentration. If uranium is present, the ratio of uranium to thorium is different in both phases.

This is shown very strikingly when the separation takes place during gelation and spheres are produced which differ markedly in colour, according to the uranium concentration.

One is reminded of the phenomenon of coacervation, but this analogy has not been worked out further.

5. — Some phenomena of gelation and washing

Many curious phenomena have been noted during this work. Since the work was directed, of course, more towards avoiding rather than studying these phenomena, mostly connected with defect particles, no exact description of conditions or explanations have been formulated.

The few photographs ([1]) accompanying this paragraph are only meant to give an impression of what is found in, unfortunately, most of the cases in which the conditions of gelation and washing are not well in harmony with the properties of the sol. All photographs refer to oxide material.

Fig. 1 illustrates the formation of droplets of water at the outside of the gel sphere in the absence of emulsifier in the organic phase (see § 2).

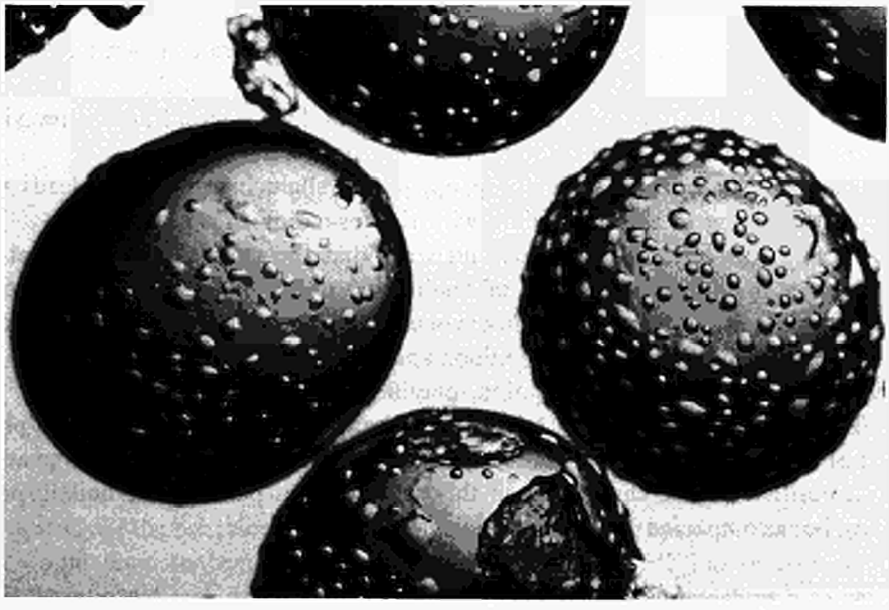

Fig. 1.

([1]) Photographs by Mr. D. J. Wanjon.

FIG. 2.

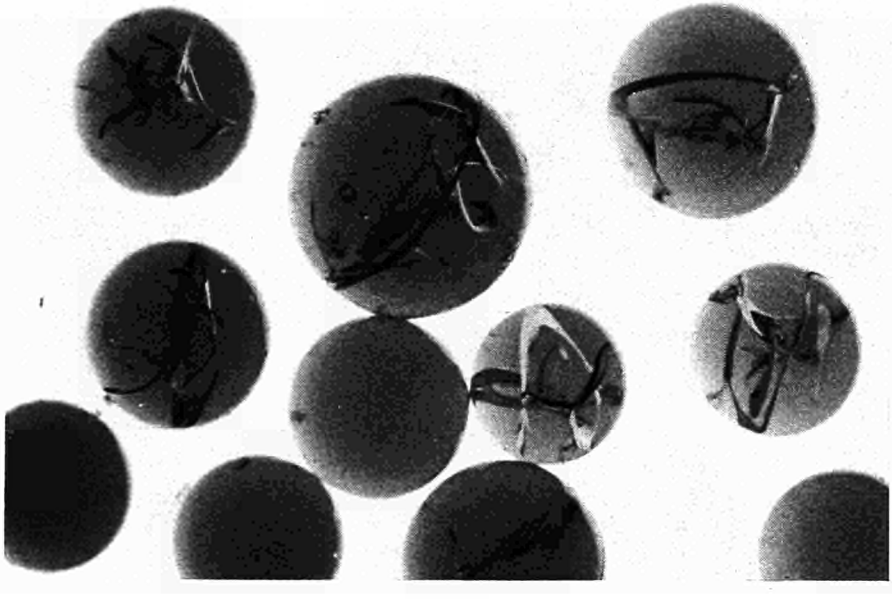

FIG. 3.

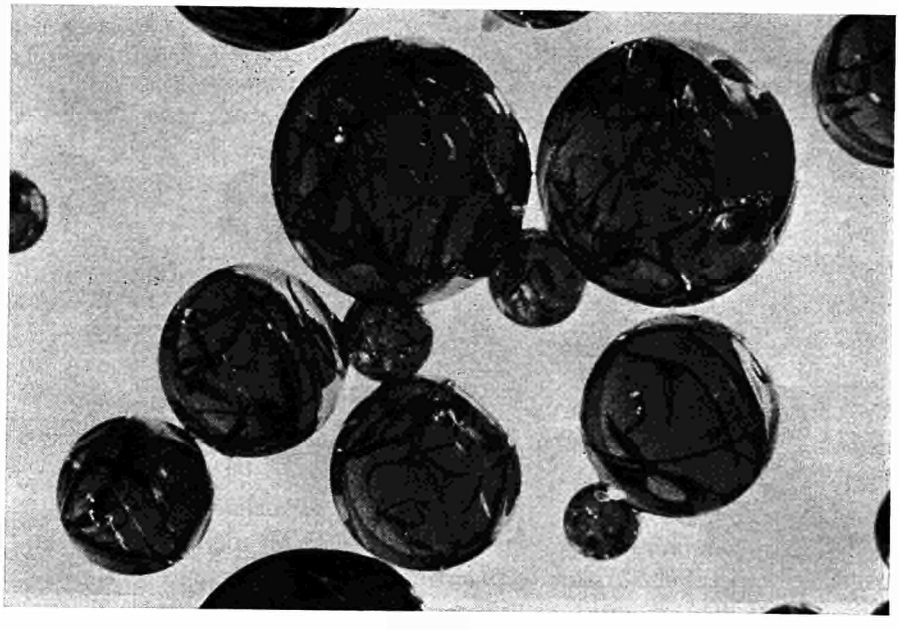

FIG. 4.

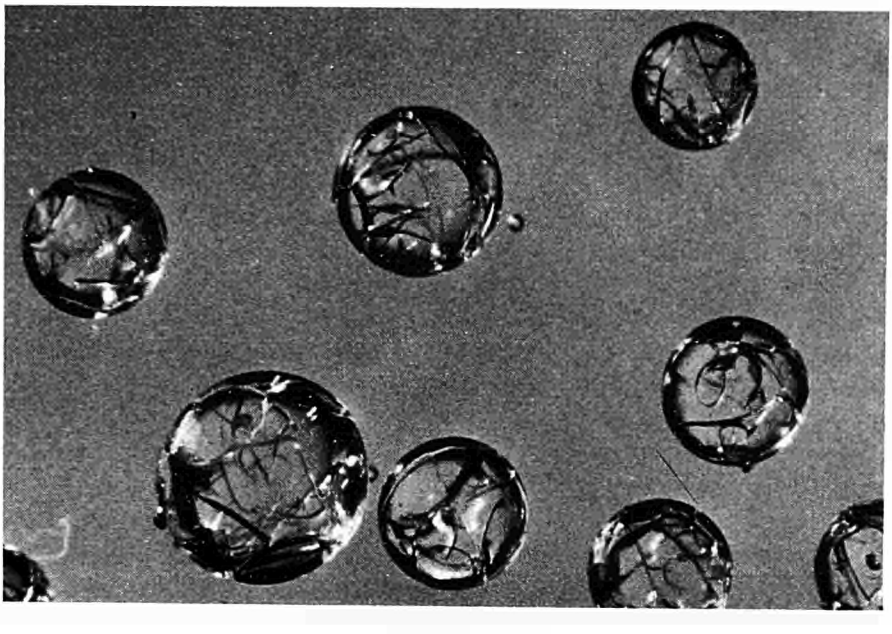

FIG. 5.

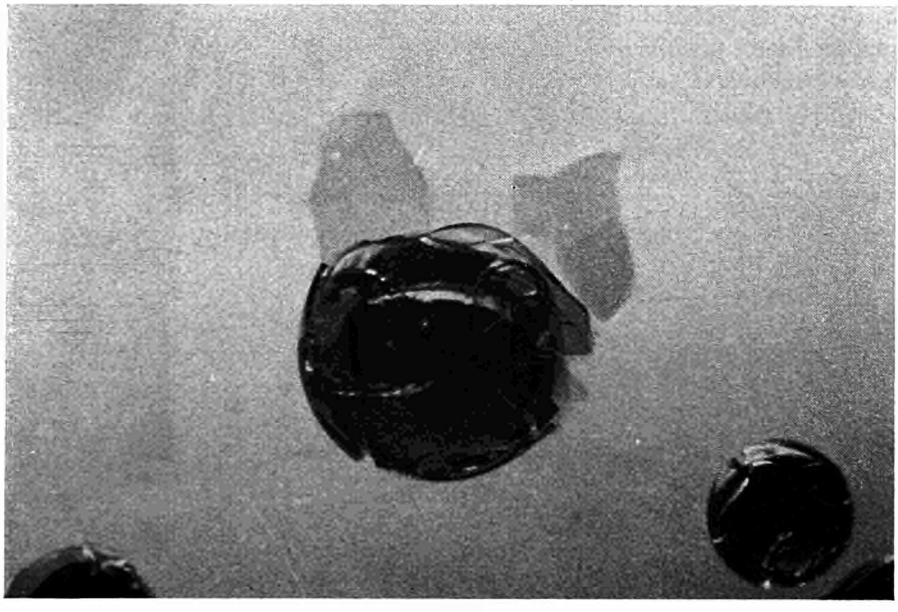

FIG. 6.

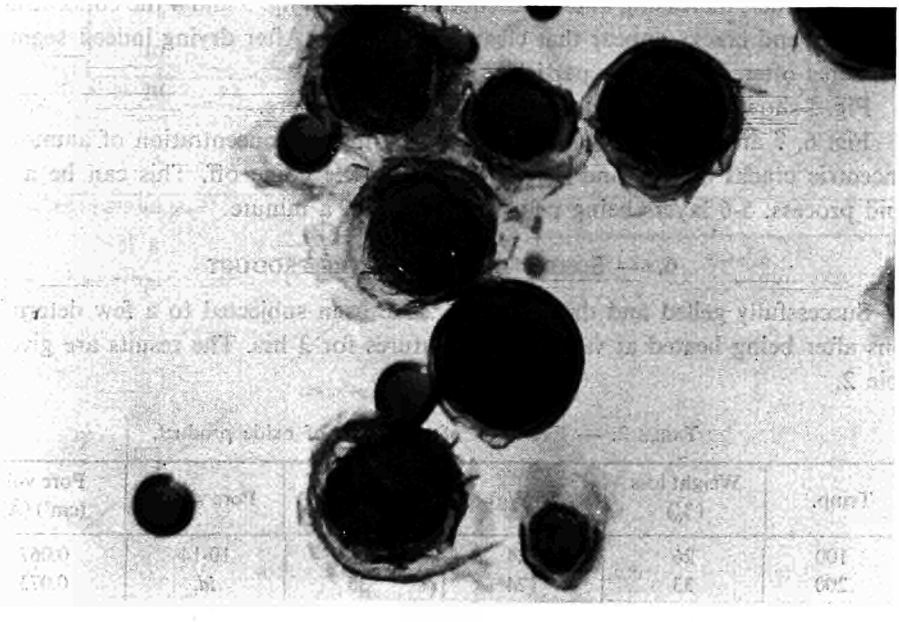

FIG. 7.

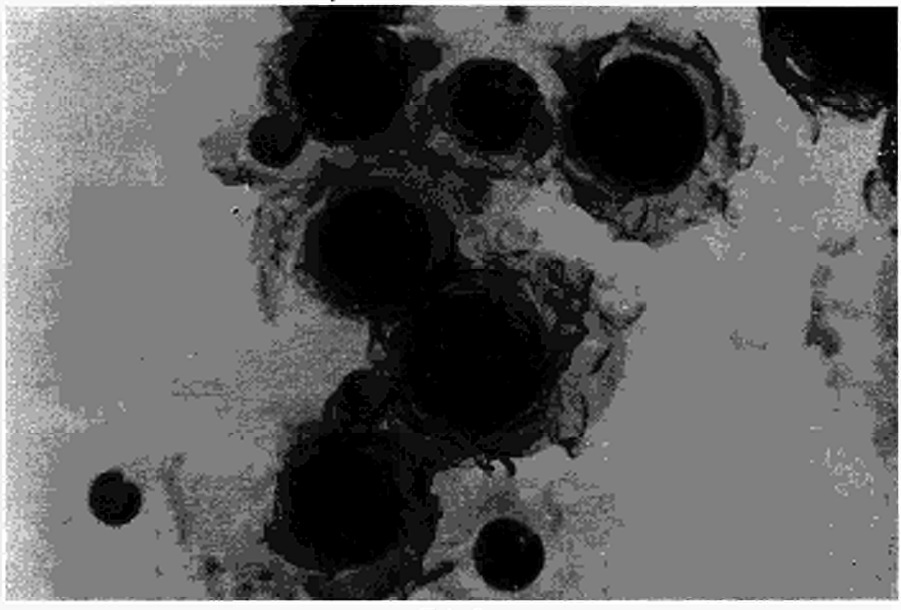

FIG. 8.

Fig. 2 shows some fragments of hollow particles sometimes found during gelation.

Fig. 3 to 8 illustrate the influence of the composition of the washing liquid. In figs. 3, 4, 6, 7 and 8 thoria-urania gelled spheres are treated with a washing liquid of, in this order, increasing ammonia concentration. In fig. 3 and 4 the concentration is too low and cracks appear that cleave the particle. After drying indeed segments of spheres often fall off the particles.

Fig. 5 shows the same defects in a gelled thoria sphere.

Fig. 6, 7 and 8 show what happens at too high a concentration of ammonia : concentric cracks appear and layers of the particle come off. This can be a very rapid process, 5-6 layers being pelt off within half a minute.

6. — SOME PROPERTIES OF THE PRODUCT

Successfully gelled and dried spheres have been subjected to a few determinations after being heated at various temperatures for 3 hrs. The results are given in table 2.

TABLE 2. — Evolution of properties of oxide product.

Temp.	Weight loss (%)	S(m^2/g)	d(Å)	Pore width	Pore vol. (cm^3) (Å)
100	26	124	35	10-14	0.067
200	33	124	35	*id.*	0.073
300	33	133	35	*id.*	0.067
1,000		13	124		0.042

In fig. 9 the full adsorption-desorption isotherm obtained for the 300° material is given. Fig. 10 gives the V_a-t plot [5] derived from the isotherm from which it can be seen that, assuming slit-shaped pores, the pore width ranges from 10-14 Å. No pores larger or smaller than these are present. The very small hysteresis loop points to the occurrence of a small fraction of pores of the ink-bottle type.

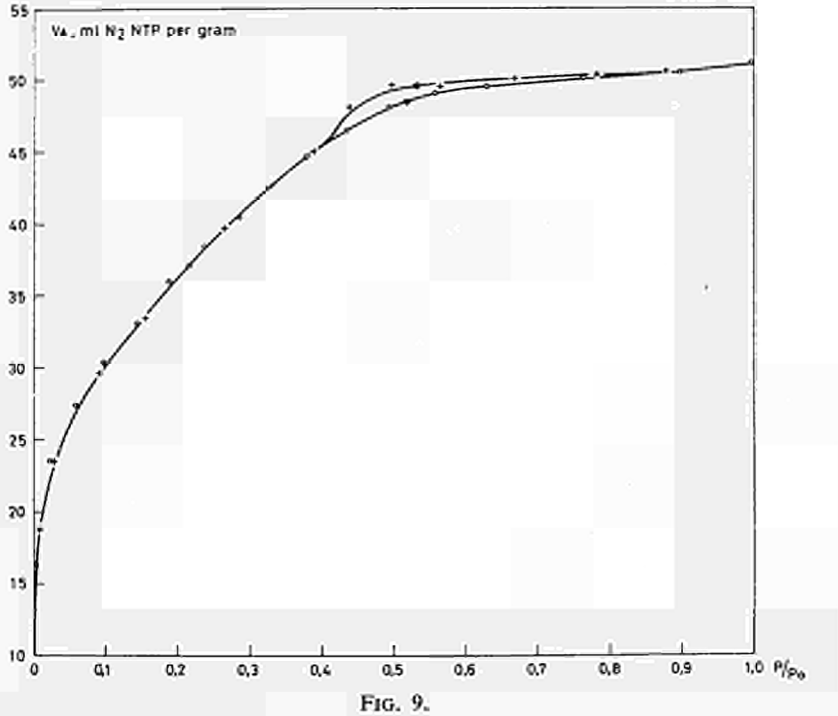

FIG. 9.

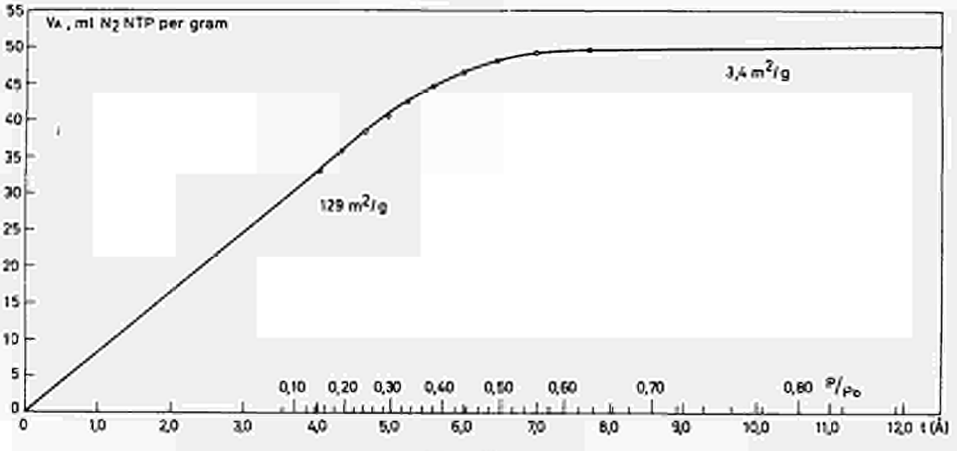

FIG. 10.

The pore volume has been obtained from the total amount of nitrogen adsorbed at $p/p_o = 0.98$, assuming that the adsorbed phase has a density equal to that of liquid nitrogen at the same temperature.

It follows from these results that the gel has an open structure, all crystallites being accessible to nitrogen. The pore volume is about 50 %. This structure is stable until at least 300° C. Somewhere between 300 and 1000° C crystal growth starts, leading to a decrease in specific surface and pore volume, but the open structure is largely preserved.

All this is in marked contrast to the behaviour of the precipiate, where low specific surfaces appear with small crystallite sizes.

ACKNOWLEDGEMENT

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SESSION II :

REPROCESSING AND REFABRICATION METHODS AND COSTS

Introduction prepared by M. ZIFFERERO, CNEN, Italy

Fuels of typical high temperature gas reactors use graphite as cladding and structural material in which spherical particles of uranium and thorium dicarbides are contained. The fuel particles (0.15 to 1 mm in diameter) are usually coated with a fission product retaining pyrolytic carbon layer. An additional coating of silicon dicarbide is present in some cases. Coated particles can be loaded as such in cavities of the graphite body or dispersed in graphite to form compacts which are then placed in the graphite log.

Several technical problems are associated with both reprocessing and refabrication of HTGR fuels : some of them are inherent to the uranium-thorium recycle and some are due to the chemical composition of the fuel. Taken as a whole reprocessing and refabricating of HTGR fuels is an entirely new problem for which existing experience on other power reactor fuels offers limited help.

Three successive steps of the processes require separate consideration :

(1) Removal of the uranium and thorium from the bulk graphite (head end step),
(2) Decontamination of uranium and thorium from fission products (purification step),
(3) Conversion of the purified products (uranium or uranium + thorium) into new fuel (refabrication step).

The combination of head end and decontamination processes is what fuel technologists call reprocessing : in the case of HTGR fuels several new factors must be considered.

These are :

(1) The large amount of graphite per unit weight of heavy metals (U + Th). Going from head end to decontamination this involves a drastic change in the plant through-put in addition to a switch from thermal or mechanical processes to purely chemical operations.
(2) The presence of the U^{232} decay chain causing a rapid ingrowth of gamma-activity in both uranium and thorium products. This leads to a variety of possibilities in the choice of the decontamination route and has determining importance in the refabrication process.
(3) The extremely high specific activity which is met during the decontamination step. This activity is due both to the high burn-ups which could be attainable and to the presence of specific hard gamma emitters such as protoactinium 233 and ingrowth of Th^{228} daughters. Severe limitations to the use of high concen-

trations and normal contactors are possible in the case of aqueous decontamination schemes.

(4) The advantage and in some cases the need of locating the refabrication facility as close as possible to the decontamination plant. In fact, quick refabrication is necessary to minimize the ingrowth of gamma-activity in the product if high decontamination has been adopted. Conversely a physical integration of the two steps is almost mandatory if the route of low or intermediate decontamination is chosen.

Foregoing considerations open at least three alternatives in plant concepts. These are :

(1) A small reprocessing and refabrication plant serving one single reactor on the reactor site. Economists can easily show, I guess, that this is the most expensive approach. It cannot be denied, however, that this solution is quite valid to demonstrate the technology and to obtain straightforwardly all the information necessary for scaling up.

(2) Use of an existing multipurpose reprocessing plant to which special head end facilities and a refabrication line have been added. With minor modifications the solvent extraction and all the necessary auxiliary services could be utilized.

(3) A single purpose, centralized reprocessing and refabrication plant, to serve a nuclear economy based on a substantial number of HTGR units totalling an installed capacity of several thousands electrical megawatts. This is clearly a target which presumes an already well established technology.

Alternative 1 and 2 are interim solutions; work in the first direction is currently going on at Oak Ridge (TURF : Thorium Uranium Recycle Facility) and in Italy (PCUT : plant for water reactors thorium-uranium fuels).

The second alternative, limited though to the reprocessing step, has also received attention at Oak Ridge whilst Eurochemic has made preliminary research on the head end problems.

A comprehensive assessment of choice No. 3 is sponsored by the Dragon : the evaluation work and, when necessary, the research and development are shared between the Dragon staff, the U. K. Atomic Energy Authority and the CNEN.

If we now examine the principal technical options for head end, decontamination and refabrication processes, taking as a basis the material presented at this Symposium, the following remarks are possible :

(1) A variety of processes has been suggested for the head end treatment of spent fuel; comparatively smaller is the number of possibilities for fission product decontamination; only two methods are considered for fuel particles refabrication.

(2) The stage of development of technology widely differs from class to class and inside the same class. Only very few methods have so far progressed beyond bench scale experiments : this is the case for the burn and leach procedure which is in course of cold demonstration in pilot plant equipment; it must be pointed

out also that an invaluable plant scale experience has been gained indirectly at Oak Ridge with the Thorex plant operation for the decontamination step and with the Kilorod program for the refabrication step.

As far as head end treatment goes, the main choice seems to be between a purely mechanical method and a high temperature oxidation method; both are followed by acid-leaching. The presence or the absence of a silicon dicarbide coating on the fuel microspheres is important in all combustion methods : due to its outstanding resistance to oxidation the coating is not attacked and the final product of the different combustion methods are the silicon carbide coated Th-U carbide particles. In this case an additional chemical or mechanical step is required to expose the Th-U carbides to subsequent leaching.

The technique of grinding and leaching overcomes in principle all difficulties arising from the presence of a silicon carbide coating : this technique has been tested successfully on a laboratory scale by the Dragon Project and was selected as first option for the evaluation of their reference reprocessing plant. According to this procedure the irradiated fuel is crushed and ground in two or three steps down to a particle size sufficient to rupture the coated microspheres.

The carbide containing powder undergoes a preliminary hydrolysis with boiling 4M nitric acid followed by a leaching in boiling 13M nitric acid. Laboratory results on non irradiated compacts have insured a uranium recovery in excess of 99,5 %. Areas needing further studies are the formation of organic matter (which could interfere with solvent extraction) and, possibly, absorption of uranium values on the bulk graphite powder.

The combustion in oxygen of HTGR graphite fuels, containing pyrocarbon coated Th-U carbides or oxides, is currently under investigation at Oak Ridge and Würenlingen. The Oak Ridge procedure has been developed up to pilot plant experiments using a 4'' diameter fluidized bed burner. According to this method chopped or crushed fuel is introduced together with alumina at the top of a burner. After heating and bed fluidization oxygen is fed from the bottom and the graphite is burned at an average controlled temperature of 700° C; the heavy metals oxides ashes and part of the alumina fall from a grid and are continuously removed at the bottom. Th and uranium are leached out completely (99,9 %) in a subsequent step with 12M boiling nitric acid — 0.04M hydrofluoric acid. The behaviour of fission products has been examined so far in lab scale tube furnace experiments on prototype Peach Bottom irradiated fuel. Results suggest that the volatilization of fission products other than noble gases will not occur to significant extent if the oxygen consumption is stoichiometric and if gases are cooled before filtration. The main problem associated with combustion in oxygen is the handling of large quantities of radioactive off-gases. A system of filters, addition of steam to off-gas followed by condensation and final filtration through absolute filters has given excellent results in waste calcination work. The Oak Ridge group is confident that a similar high efficiency cleanup system can be devised.

Results on the combustion of graphite fuel element as such (without chopping) in a high frequency induction furnace are given in the paper by Züst and coworkers of Würenlingen.

Laboratory scale experimental results on oxygen combustion are presented also by the Eurochemic group. An alternative combustion with nitrogen dioxide has shown that oxidation reaction initiates at lower temperatures in this case and that acceptable rate of combustion can be obtained at 400° C when oxygen combustion is inefficient. At higher temperature, however, (600° C) the oxygen combustion is faster. Advantages of lower temperature on fission product volatilization must be weighed against nitric oxide corrosion, reaction time and increase in the off gas volume. The CEN group, headed by Dr. Schmets, is reporting similar results on the carbon oxidation with nitric vapors : the addition of oxygen to nitric oxide increases the reaction rate.

Bench scale work on anodic disintegration of compacts is carried out (on behalf of the Dragon) at CNEN. Results so far obtained indicate a technical possibility of separating the coated particles from the carbon matrix of the compact. Evidently a prerequisite of this route is the possibility of an easy access of the solution to the compact.

If fuel particles are coated with a silicon carbide layer the final product of combustion or anodic disintegration is the particle still covered with the coating. Since silicon carbide is not removed by leaching agents a further step is required to expose the U and Th carbides. This could be obtained by grinding. An alternative to grinding is suggested by Eurochemic consisting of a high temperature (600° C) treatment in a molten bath of alkali carbonates with oxygen, air or nitrogen dioxide supplied to the melt by sparging. In these conditions silicon is removed as a silicate and a dilute solution of nitric acid can be used to bring in solution all the melt.

The principal options on the fission products decontamination process are focused in the paper contributed by the Dragon Project. The Project has decided to perform assessment studies on two basic reprocessing philosophies, namely intermediate decontamination of both uranium and thorium and high (or maximum) decontamination of the uranium; thorium being stored separately from wastes, for a period of time long enough to allow the decay of gamma-active daughters of Th^{228}.

The uranium and thorium products obtained by the first route will achieve a decontamination factor in the order of 10^4 in a single codecontamination cycle. For the reference fuel under examination the residual gamma-activity should be in the order of 0.3 curies Mev per kg of U + Th. This activity level is sensibly equal to the gamma- activity of the U^{232} decay chain in the refabrication process. The refabrication of the coextracted U and Th products would be performed in remotely operated heavily shielded facilities.

The high decontamination alternative should permit refabrication of recycle uranium (and virgin thorium) in lightly shielded equipment. In this case the purification process is based on a three cycle solvent extraction. The bulk of fission products

leaves the codecontamination cycle in the raffinate; thorium is left in the aqueous raffinate of the second cycle and the uranium stream undergoes a third purification cycle. The decontamination factors required for the reference fuel are in the order of 10^6. Expected D. F. should reach 10^7 with a final activity of approximately 2 mc Mev per kg or uranium.

Used TBP solvent undergoes an alkaline wash and a steam flash distillation before being recycled. This precaution seems adequate to minimize the effects of the highly active feeds. Solvent extraction schemes seem to necessitate a limited development work since a wealth of information is available from the Thorex program.

A potential non-aqueous alternative for fission product decontamination is given by the fluoride volatility process. Work in this direction is mentioned in the paper by Nicholson and coworkers and reported by CEN group. The process seems to be applicable to those systems in which, after the head end treatment, uranium and thorium are contained as oxides. According to the Dragon paper in fact silicon carbide coated fuel particles should undergo grinding and combustion before being fluorinated due to the presence of an inner pyrocarbon coating which could cause explosions initiated by fluorine carbon compounds. This would complicate unnecessarily the process. Fluoride volatility applied to oxides containing material is limited on the other hand because it is unsuitable for the recovery of thorium from non volatile fission products. This causes a substantial increase of costs for high level wastes storage. Preliminary experiments at Oak Ridge indicate also that the combustion fluorination of fuel containing Th-U oxides microspheres prepared with sol-gel will not be applicable due to the high resistance of the material to fluorination. Information on high temperature reaction of uranium dioxide and dicarbide with sulfur hexafluoride to yield uranium hexafluoride are contributed by Laser and coworkers of Jülich. Based on this reaction a reprocessing scheme is suggested; an interesting feature is the non corrosive and non poisonous properties of the basic reagent up to 500° C.

The last aspect to be considered is *fuel refabrication.* A distinction must be made between refabrication mode and refabrication process also if there are some technical and economic dependencies. The mode of refabrication can be of the contact type (direct, with hoods or with glove boxes), of the semiremote type (requiring light shielding) and of the remote type (under heavy shielding). In the case of uranium-233 recycle the mode of refabrication depends upon the degree of decontamination, the U^{232} concentration, the age (i.e. the time interval elapsed from decontamination), the quantity and whether virgin or recycle thorium is used.

It also depends upon the margin of protection desired against off standard conditions, such as occasional rework, equipment failure or human error. The possibility of a contact type refabrication of recycle uranium 233 is highly questioned : the almost general agreement is that, for U-Th recycle, at least semiremote refabrication must be envisaged, due to the equilibrium concentration of U^{232}.

The refabrication process itself offers a limited choice : papers on this subject are submitted by the Dragon Project and by the Oak Ridge group of Lotts and

coworkers. For sake of presentation a refabrication process can be divided into three steps :

(1) Preparation of uranium and thorium carbide microspheres,
(2) Coating of the microspheres,
(3) Preparation of compacts and fuel element assembling.

Essentially two routes for particle preparation are in course of evaluation at the Dragon Project : these are a powder metallurgical method and a sol-gel process. Experience at Oak Ridge has already demonstrated on a pilot plant basis a sol-gel process. Dragon is confident, on the other hand, on the possibility of remotizing the powder metallurgical process adopted for the contact fabrication of the first core particles.

Block diagram steps of the powder metallurgical approach include : dry mixing in the appropriate proportion of thoria, urania and carbon ; granulation of the consolidated material obtained by pressing or extruding the mix after addition of a paraffin binder; granules spheroidization by abrasion in a planetary mill. After sieving, proper mesh-size particles undergo carbothermal reduction by heat treatment in vacuum at 2,300° C. According to Dragon experience the process bottleneck of this method is the granulation step. Sol-gel development work is carried out on behalf of the Dragon Project by the KEMA group at Arnhem and by the group of Cogliati at CNEN, Rome. Sol-gel was first developed at Oak Ridge for the preparation of high density Th-U oxides. The foundation of the method is well-known : a colloidal (sol) solution is prepared which contains thorium and uranium in the form of highly hydrolized nitrates; it practically consists of concentrated hydroxides with a low nitrate ions content. Microspheres are prepared by dispersing the sol droplets in an immiscible organic liquid which has some solubility for water. In this way water is removed from the sol droplets causing gelation. Particles are then collected, dried and calcined to yield a high density material. If carbon is added to the sol, carbon containing oxides spheres are prepared that can be converted to carbides by an appropriate high temperature treatment. Alternatively oxides microspheres can be converted in carbides by high temperature treatment with carbon powder as envisioned in the Oak Ridge paper by Lotts and coworkers. The methods for the preparation of a stable sol and the gelation of processes under development at KEMA and CNEN are given in the Dragon Project paper by Horsley and coworkers : further information on process chemistry is contained in two additional papers.

The KEMA process appears to be suitable for refabricating uranium and thorium carbides when uranium and thorium are received in separate streams from the reprocessing plant. When uranium and thorium are not separated in the decontamination process the CNEN method is considered the most appropriate; as pointed out by the Dragon experts, this method would also facilitate the recycling of reject material since a simple reconversion to the combined uranyl-thorium nitrate is sufficient.

It is maybe worth mentioning that all foregoing particle preparation methods can be applied with minor modification to the fabrication of oxides.

Particle coating is a delicate step and represents an important factor in refabricating economy : development work is necessary to bring this process in the production stage at lower costs. Both particle coating and the preparation of compacts, this last does not present particular problems, are independent of the mode of particle preparation.

A comprehensive program directed towards the experimental assessment of the different HTGR fuel refabrication steps under remote conditions on pilot plant scale will be carried out in the Thorium Uranium Recycle Facility at Oak Ridge. This facility will be capable of processing and refabricating HTGR fuels up to 12 feet long at a daily rate of 35 kg of Th-U irradiated to 25,000 MWD/t and decayed for 90 days.

Once processes and equipment will be tested and operated in the TURF, a sound basis for the cost evaluation of HTGR fuel processing and refabrication will be available. So far this portion of the fuel cycle cost is difficult to assess and cost figures presented at this Symposium are admittedly highly speculative. Long lists of assumtions are necessary to make a comparison possible. Talking of cost comparison one should always bear in mind that evaluation of fuel cycle costs for second generation reactors should be compared with projected costs of present reactors. This adds second order uncertainties.

As far as reprocessing goes, a good starting point for comparison is the existence of a commercial reprocessing plant now close to the start-up, built by Nuclear Fuel Service.

Based on plant nominal capacity and service rates, a reprocessing cost corresponding to 0.2 mills/kWh (electrical) can be extrapolated for water reactor fuels (BWR or PWR type). These are projected costs for 20,000 MWD/t burn-ups and include only the reprocessing operation. However, when necessary, NFS capactity could be increased with minor investments and this could lead to a reduction in the reprocessing charges.

Projection of fuel fabrication costs for water type reactors is maybe more difficult. Assuming the same 20,000 MWD/t burn-up and a projected fabrication cost of 60-80 $/kg of fuel, fabricating costs would correspond to approximately 0.4-0-6 mills/kWh (electrical).

HTGR reprocessing and refabrication costs evaluation is included in the assessment study initiated in 1964 by the Dragon Project : accurate predictions are not expected from this study, but it was considered that cost estimates sufficiently approximated to be used in optimisation studies were possible. It was assumed that the reference plant would include reprocessing and refabrication and serve a program of 10,000 MW (thermal) of HTGR reactors. The reference fuel would have a 150,000 MWD/t burn-up and would contain annular fuel compacts in a hexagonal graphite lattice. Technical options on head end treatment, decontamination and fuel fabrication have been mentioned earlier in this introduction. Work on this assessment is in progress but order of cost estimates for reprocessing and refabrication will not be available unfortunately before the end of 1966.

Separate cost estimates for reprocessing and refabrication are contained in two Oak Ridge communications allocated to this session.

A preliminary economic evaluation for a head end facility based on the burn and leach method was made by Nicholson and coworkers. This facility includes reception and storage, crushing, burning and leaching, feed adjustment, feed storage surge capacity and tank storage for recovered thorium decay. Plant daily through-put is approximately 4 tons of graphite, 400 kg of thorium plus uranium, 36 kg of fission products. This through-put corresponds to the fuel discharged by an installed thermal capacity of 25,000 MW(th). Further assumptions are : 225 days a year operation on fuel burnt up to 50-80,000 MWD/t, 6 months cooled. The reference fuel is made by 20 feet long graphite tubes in which pyrocarbon coated oxide particles are contained. The estimated capital cost of this facility is 9 million dollars; running costs when the plant is in "stand by" amount to 500$/day. In full scale operation total running costs reach 1,900 $/day. As pointed out by the authors, the translation of these figures into unit costs depends critically on the plant loading and on the limitations imposed by the associated solvent extraction plant. When operating at full capacity the head end facility could process fuel up to the feed for solvent extraction at unit costs corresponding to 0.04 to 0.06 mills/kWh (electrical). Based on a superficial comparison of building and equipment size with NSF the authors estimate that it should be possible to build and operate a single purpose reprocessing plant with head end and solvent extraction having the above mentioned capacity of 90 tons/year at a cost which is not greater than those of NFS. Assuming the same type of financing such a plant could charge $ 75 to $ 85 per kg of heavy metal corresponding to a unit cost of 0.11 to 0.15 mills/kWh electrical. This is indeed a very attractive figure but it is doubtful if the required fuel load will be available in the next 15 to 20 years. Small reprocessing plant for specific fuel elements should be taken into consideration as interim solutions. If fuel particles could be easily separated from the graphite hardware small size head end would be required; the use of a single one cycle codecontamination flowsheet suitable for remote fabrication could reduce heavily capital and operating costs. Work in this direction seems to deserve more attention.

Extensive refabrication cost analyses are reported in the paper of Lotts and coworkers from Oak Ridge. They result from a computer program set up at Oak Ridge in order to obtain a complete analysis of the refabrication costs for any reactor fuel element, including HTGR elements. The number of alternatives discussed and the extended range of variables considered make this evaluation useful for the refabrication of virtually any fuel element for high temperature gas-cooled reactors.

Basic assumptions of this analysis are the following :

(1) Starting material is in the form of oxide microspheres (either separate UO_2 and ThO_2 or mixed oxides); the fabrication costs of spherical particles by the sol-gel process are anyhow mentioned.

(2) The refabrication flowsheets involves two alternatives since uranium and thorium carbides can be manufactured separately from the two oxides or together from a mixed oxide.

(3) Four different values of plant capacity ranging from 60 to 3,700 Kg of heavy metal per day are considered.
(4) The refabrication process was divided into three zones : conversion of oxides to carbides, particle coating and compact preparation plus fuel assembling.

The material location, quantity and age were calculated for the different zones.

Based on the foregoing assumptions the degree of shielding required was plotted against the U^{232} content in heavy metal. Then, the following cost determining parameters were explored :

(1) Effect of mode of fabrication (contact, semiremote, remote);
(2) Effect of recycle method (recycling of uranium and thorium);
(recycling of uranium)
(recycling as mixed carbides)
(recycling as separated carbides);
(3) Effect of fuel composition (oxides versus carbides);
(4) Effect of U^{232} concentration.

It is practically impossible to condense in a few words the results. Some conclusions (or more appropriately trends) of great importance must be mentioned : shielding calculations show that if a U^{232} concentration in the order of 50 ppm is expected, in equilibrium cycle fuel, a remote plant is required for refabrication. Contact fabrication of virgin thorium carbide coupled with remote refabrication of recycle uranium offer costwise very little advantages when compared with remote refabrication of recycle uranium and thorium or remote fabrication of recycle uranium and virgin thorium; this is true even though, in the first case, a substantial quantity of fuel is not fabricated in a remote plant.

Incremental costs from oxides to carbides range from approximately 15 % at low production rates to 5 % at high production rates. The coating operation (limited to one single layer of pyrolytic carbon) accounts for 30 % of the total refabrication costs at low capacities and 40 % at high capacity. This fact should induce from one side fuel elements designers to a very careful use of the multiple coating philosophy : on the other side metallurgists should strive to increase coating efficiencies.

Predicted costs for a typical recycle HTGR element as a function of plant capacity indicate that a refabrication plant designed to serve a program of 10,000 thermal MW spread over several HTGR reactors could refabricate the fuel at an estimated cost of 300 to 350$/kg of contained heavy metal. In the hypothesis of very high burn-ups (100,000 MWD/t) this would correspond to about 0.30 to 0.35 mill/kWh (electrical).

In conclusion, fuel cycle costs for high temperature gas reactors are founded today on a rather limited technological background. First results are encouraging in that the cost penalties of recycling fuel do not promise to be unbearable; much development and assessment work is still required. The operation of facilities specially devoted to the thorium uranium cycle such as TURF and PCUT will undoubtedly give a better basis to the technology and the economics of these fuels.

SESSION II : DISCUSSION

Chairman : P. CAPRIOGLIO (Euratom)

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1. — AUTHORS' COMMENTS.

Dr. CAPRIOGLIO : I should like to follow the same method we followed this morning, and now ask the authors to make their comments or their further presentation, if they wish so.

Dr. WOOD *(Dragon Project)* : I would like first to make a few general remarks on the philosophy underlying the Dragon study and secondly to bring you up to date on some of the information which are given in the paper. As you will appreciate from the paper, Dragon Project is not directly engaged in carrying out experimental reprocessing and refabrication studies on HTR fuel, and I must emphasize the *re*-processing and *re*-fabrication, because we are of course doing a lot of work in the fabrication field. The Project's decision to abstain from comprehensive participation in the experimental field on reprocessing was, I think, realistic, because it is clear that at this stage of the Project's life, budget provision would be unlikely to be adequate. It is a costly field. However, our interest in this field is obvious, hence it had to be decided what work could be done on a limited budget. It was considered that a reasonable return was likely to be obtained from a detailed engineering assessment of the costs of reprocessing and refabrication, a field of prime importance in fuel cycle optimization studies, but one in which authenticated cost estimates inevitably are lacking. The next decision was to select the head end and reprocessing route to be used in the study. The variety of head end processes described in the papers submitted today is, I think, in itself sufficient evidence that no single process has yet been found to have clear superiority over all others. In a round about way, this lack of a clear winner made our choice somewhat easier. We decided that we would use existing technology wherever possible, for example in the selection of the Thorex type decontamination process; but on the other hand where the technology has not been demonstrated, we specified the simplest or apparently simplest of the many alternatives that had been proposed. This led us to the choice, rightly or wrongly, of the grind-leach route for the head end process. Now, of course, we fully admit that we can be wrong in our choice because the apparently simplest may turn out in practice to have unknown complications. Nevertheless, we think that at this stage of HTR fuel cycle development, the principle of choosing a fairly simple approach for detailed examination is correct, even though ultimately it may be necessary to introduce modifications. Hence, we make no apology if our choice of process appears to be lacking in sophistication or elegance in comparison with some of the more exotic routes proposed. Even so, we have tried to have the best of both worlds by building into the process specification at selected point,

certain refinements which may facilitate the treatment and cheapen costs. The electrolytic modification of the head end process is a case in point. We are well aware of course that confirmation of the choice of process route and the cost estimates which we hope will come out of the study, can only be obtained by a detailed and substantial development programme such as Oak Ridge will carry out in the TURF facility. Although similar provision cannot be made in this particular study, nevertheless we hope that we shall at least have narrowed the range of experimental work by eliminating a few of the present areas of uncertainty. I should like to acknowledge here the very great contribution which our collaborators at CNEN and in the UKAEA have made in this particular study.

Finally, in bringing up to date the information given in the paper, I can say that a few simplifications have been introduced since the paper was written. These are as follows :

First, the scrubbing of the off gases from the grinding section of the head end process has been eliminated; we believe that filters alone will be adequate to cope with the off gas decontamination problem.

Secondly, the preliminary hydrolysis of the crushed fuel with 4 molar nitric acid has been abandoned, because we think that if adequate control of the feed of the material can be achieved, and we think it can, then we can feed directly into 13 M nitric acid.

Thirdly, the long refluxing with nitric acid has been abandoned because we think there is no significant gain in the degradation of the organic products by prolonged boiling.

Mr. NICHOLSON *(Oak Ridge)* : I have only a few comments. First, to correct a small error in the report : on page 153, we indicated that boiling in the bed did not interfere with the leaching. Actually, we could tolerate only a small amount of boiling before there was difficulty in entrainment of the solids with the leaching liquors. As far as further experimental work is concerned, we have done larger scale leaching tests with ash from burning of Peach Bottom type fuel compacts. We find that with a very limited amount of washing we have reduced the uranium losses to less than 1/4 %, so that we hope that ultimately, the losses in the leaching operation after burning will be essentially zero.

One other point concerns the stability of mixed uranium and thorium oxide particles to high temperature oxidation. We find that particles containing as much as 75 % UO_2, and 25 % thorium oxide are not appreciably attacked by hot oxygen at 700 to 750° C. Now, as far as a reason for the choice of our burn-leach route, we had some earlier experience with a different type of graphite fuel. The preliminary experiments were not very encouraging for grinding and leaching and getting complete recovery of uranium. With reference to the Dragon fuel, I believe that the grinding and leaching is a very logical choice, because after all, the silicon carbide particle is rather refractory to most aqueous reagents. So we have no argument at all with them on that.

Mr. LOTTS *(Oak Ridge)* : I have only several brief comments. First I would like to emphasize that the cost analyses are based on speculation about mature technology and they are not based on any significant experience.

Secondly, I would like to emphasize that there are several objectives in selecting the process to be used for refabrication. First, and I think this has been emphasized already, the process should be as simple and as short as possible. Secondly, it should be easily automated; we do not believe that it is practical to consider the use of master slave manipulators in refabrication of such fuel. Thirdly, one should try to minimize the recycle material within the process. I would cite the Kilorod experience in this case as a procedure, in which we had very little recycle material within the process. Essentially 100 % of sol-gel material was loaded directly into the fuel rods. This should also be the objective in the case of HTGR fuel fabrication.

The other thing that I would like to clarify, possibly this is a misunderstanding on my part, we have not had experience with the remote preparation of sol-gel microspheres. The experience in sol-gel on significant scale remotely has been that of the preparation of shards of oxide material. We are however now preparing a facility for microspheres, where we will attempt to prepare as much as 10 kg in an 8 hour-shift in a single column.

Dr. LOPEZ-MENCHERO *(Eurochemic)* : Concerning the carbon attack step which could be considered as common to any European carbide fuel, like AVR or Dragon, we feel that the method is combustion and we have practically nothing to add to the work of Oak Ridge, except that the use of nitrogen dioxide might lead to lower temperatures which would possibly influence the reactivity of the oxides resulting from the combustion of the carbides of the fertile material. Concerning the specific case of Dragon, silicon carbide coatings, we would like to say that the molten salt process which we describe is *one* way out but not *the only* way out.

I have to mention that when we started to do this work, the Dragon people had already started their grinding method. There was an agreement that because they were following that line, we would try other ways out. So, not considering crushing, we did not find any other way out than this attack at rather low temperature with the eutectic of the alkali metal carbonates. To talk about cost of this process would be premature, because this work was stopped one and a half year ago; we can only say that the waste oxide that would be produced per kg of uranium treated is not much higher than from MTR fuels in conventional aqueous processing. But if of course there is a possibility, like the crushing method, of attacking the silicon carbide coatings without producing a supplementary waste, and the operation is not costly, then we have nothing against that; it is just another way out. Apart from the fact that crushing was already being studied, we considered that a molten method could be approached on a laboratory scale, but that the crushing method would give only significant results if it were studied on a rather bigger scale. That a particle can be crushed is obvious; that the carbides of the fertile elements

can be hydrolyzed is also obvious; the problem is whether this grinding on a full scale is less expensive than any other method.

Dr. CAPRIOGLIO : Thank you very much for your rather challenging addition. I would like to ask now Mr. Laser.

Dr. MERZ *(KFA)* : I do not think we have to add anything at the moment to what is written in our paper.

Dr. BAERTSCHI *(Eidg. Inst. f. Reaktorforschung)* : Dr. Zifferero has mentioned the method which we have used at Würenlingen for the treatment of high temperature carbide fuel and I shall give a short summary and some additional comments here.

Our treatment is based on the combustion of the whole carbide-graphite fuel elements in oxygen. Heat is applied by high frequency induction in a specially designed water-cooled vertical tube furnace which is schematically shown in Fig. 4 of our paper. In our case, this furnace is long enough to contain a whole Dragon-type fuel rod, its design, however, could be adjusted to a wide variety of fuel elements ranging from whole fuel bundles to pebbles. The lower end of the furnace, as shown in Fig. 5 is equipped with a water cooled grate and a mechanical device to break up loosely sintered pieces of ash. The off gas decontamination system consists of a counter current scrubber column, packed with stainless steel wire gauze rings, through which percolates a solution of sodium bicarbonate. In our present set up we use in addition an electrical precipitator in order to remove any carry over from the scrubber column. Further runs with high active material will show, however, whether this electrical filter will be necessary or not in a technical plant. The furnace and the scrubber column are operated under slight vacuum and some air entering the upper part of the furnace prevents the formation of carbon monoxide. In the present installation we achieved an average burning rate of some 0.75 kg of fuel element per hour and it seems possible to increase the throughput by a factor of about 10 in a properly designed bigger furnace. In succeeding runs, a yield of fuel ash close to 100 % was obtained, since the holdup of ash in the furnace is only a few grams and the carry over of light ashes into the scrubber is very low (0.2 %). Tracer experiments with slightly irradiated uranium/thorium carbide fuel showed about 0.1 % of the total activity to be present in the scrubber solution and no detectable activity leaving it. We consider this low carry over of fission products and uranium as one of the advantages of this burning method.

Experiments with coated particle fuels showed, that pyrolytic carbon coated fuel was readily decomposed, while silicon carbide coated particles were hardly affected. But also in this latter case the method appears to be very useful concentrating large amounts of high active material without laborious and expensive procedures. In the near future we shall start experiments with high active Dragon fuel which has been irradiated in the DIORIT at Würenlingen to a burnup of about 14 %. From these experiments, we will get more information on the performance of the off gas treatment system.

Dr. SCHMETS *(CEN Mol)* : I have some comments to make. We have collected data obtained on UO_2 and UC fuels, and we are not specialists in the uranium/thorium fuel cycle. But the data collected are related somewhat to the object of the present symposium. In our paper, we propose a new way to process pyrocarbon coated fuels, and what we propose is to use nitric acid vapour. The data we have collected indicate that rapid reaction can be obtained. All results seem to be somewhat different from Eurochemic results, in that sense that we have, apparently, satisfactory rates at lower temperature, and without corrosion. The corrosion data obtained are for example for Nionel 0.125 mgm per sq cm for a thousand hours operation at 375° C. An important fact also is that the rate of reaction is increased by oxygen and I think this is not related to the presence of oxygen itself, but to the shifting of the equilibrium of the nitrogen oxides. One advantage of the method is the increasing chemical activity of the obtained oxides.

I think that the effluent gas treatment for this system should be also commented : as far as we were able to go at the bench-scale, this treatment will be very similar to an aqueous plant gaseous effluent treatment.

For the technicological development of the proposed process, I think it is too early to make a decision between aqueous and dry methods for the uranium recovery step. If aqueous method is used, the leaching could be performed in the same vessel. It is difficult to answer the same question if the fluorination method is used after oxidation; corrosion problems associated with the use of strong oxidating and fluorinating agents could be important. We have made some tests with a mild fluorating agent (SF_4). The corrosion test indicated that the same vessel could be used for alternate use of nitric acid vapor and SF_4.

There has been an objection against the fluorination process : it is the high density of uranium thorium oxide which could inhibit the uranium fluorination. It is too early to decide anything about that because the density and the physical state of the particles to be reprocessed will be quite different after irradiation.

The last point I want to mention is reconversion possibilities, if the final product of reprocessing is uranium hexafluoride.

2. — INTEGRATED OR CENTRALIZED PLANT.

Dr. CAPRIOGLIO : Any further general comment? Well, if not, we will open the discussion. There is one thing that I am particularly interested in, in the presentation by Dr. Zifferero, this is the difference between an integrated plant and a centralized plant. If I am not wrong, he said that there is not very much hope for an integrated plant at long term, and that it would only be of interest for the moment, since obviously the market is not large enough. I hope I got it right. If this is the case, is everybody of his opinion, and is there no serious incentive to have integrated plants, in particular if one wants to go to very high conversion factors and possibly breeding? In this case perhaps the advantage of having an integrated plant allowing for quick reprocessing would be perhaps essential. Is there any feel-

ing on this from the authors; I would like to start from Dragon again. No? No specific opinion from Dragon. May I ask Oak Ridge? Is Mr. Nicholson wishing to comment on this?

Mr. NICHOLSON *(Oak Ridge)* : I do not believe I have any comment either. I think the facts of integrated vs. large central plants are pretty self evident.

Mr. ROSENTHAL *(Oak Ridge)* : I would like to comment by noting that people disregard the possibility of fabricating and processing on site because they assume that the fuel elements and the procedures for handling them, and the reactors are going to be as they are described today by the installations which are developing HTGRs. I suggest that in the future it may well be that the fabrication reprocessing scheme will dominate the design of the reactor, and that one then might find it quite attractive to use fully integrated processing and refabrication in order to achieve what Mr. Caprioglio suggested, possibly a breeding system, and that this particular idea looks only unattractive when one starts with the assumption that he knows what the core of the reactor looks like, and I make this as a challenge to reactor designers, and perhaps we ought to start at the other end sometimes and we see where we end up.

Dr. CAPRIOGLIO : I do not assume, any reactor designer would like to take the challenge up now, but if there was any...

Mr. HOSEGOOD *(Dragon Project)* : It may introduce interesting possibi'itites if we start trying to design reactors around the processing plant rather than the processing plant on the reactor site. However there is a small point, apart from the scale effect which is usually so important in the cost of such plants; the continuous use of this expensive capital equipment has also a very strong influence on the unit cost of the operation. If we have processing plant serving one reactor on the reactor site, then we have to adopt a fuel charging and discharging scheme with the reactor, which will maintain a continuous uniform load of fuel being reprocessed and refabricated on the plant, without involving very large stocks of fuel being held on the site.

Prof. SCHULTEN *(KFA/THTR)* : As a reactor designer, I would like very much to have an integrated reprocessing system on the site, but we are not so very well informed to have enough knowledge what are the real costs of such a kind of system. In our case, we believe that we can make a continuous loading and unloading of the reactor. Costs of about 9 or 10 million dollars for a reprocessing plant of a size of 200 kg per day would be acceptable for a 1,000 MW reactor. And so, without having any knowledge about it, I believe that when reprocessing plants can be built in the order of about 10 million dollars, then it is not quite excluded that integrated systems connected with 1,000 MW reactors perhaps can be built.

Mr. NICHOLSON *(Oak Ridge)* : I believe the figure of 9 million dollars, quoted as representative of the cost of a reprocessing plant, is just the cost for adding a

head end facility to an existing reprocessing plant in our study. It provides the major share of the reprocessing capability; admittedly, this is the cost for a rather large head end added to an existing facility. I do not have much doubt either that a very small, complete reprocessing facility could probably be built for less than 38 or 39 million dollars, but I would hate to even guess a figure. The basic problem with these much smaller plants is that the unit cost of manufacturing and reprocessing the fuel would be quite high. This is again the effect of scale.

Dr. SHEPHERD *(Dragon Project)* : I think we ought to be rather careful about thinking too much in terms of integrated reprocessing. Possibly, one of the attractive applications for the high-temperature reactor will be in smaller units, as well as bigger ones, and these would certainly not be able to operate economically with integrated reprocessing facilities. So I think even if one did integrate reprocessing with the very large plant, one would still want some central reprocessing facility to deal with fuel from a number of smaller reactors.

Dr. CAPRIOGLIO : I take the points; I am only wondering what would be considered to be a small unit in twenty years from now, because the evolution of the production of electricity is so fast in this field, that I am sure that by that time what we call now small power stations would probably not exist any more in practice, in Europe or in the United States, but I may be completely wrong.

Mr. NICHOLSON *(Oak Ridge)* : My comment was not on trying to predict what a small plant would be in 10 or 20 years. In course of making this study we have in our paper, it became quite apparent that a very significant cost was attached to the fact that we had to ship, store and handle a very large and bulky fuel element — 20 feet long elements are difficult — and I want to emphasize one of the conclusions that was mentioned in our report. If the reactor designer can design a fuel element with separable particles, so that the used log could be either discarded at the reactor or perhaps even reloaded with fresh fuel particles, if this is feasible, then the reprocessing plant would have to work only with the highly concentrated fuel particles. You would find that the fuel shipping costs and storage and handling costs for those particles would be far lower, particularly if you have an appreciable distance from the reprocessing plant to the various reactors.

Dr. CAPRIOGLIO : I think that you are making now a plea for loose coated particles that we discussed this morning. I think that this is in fact the most obvious advantage of loose coated particles, from what I gathered this morning, and I do not think, on the other hand, that we should take up the discussion again. I am sure that fuel element designers are aware of this problem, and once they will start making calculations for the integrated fuel cycle course, they will realize how much the transportation is coming into their assessments.

DIPL.-ING. MULLER *(BBK)* : I think a short remark on this integrated plant question should be made. If we look into the advantages of the high temperature

reactors, one of the big advantages is having a reasonable high specific power. I would like to refer to Dr. Stewart's paper which he is discussing tomorrow, where he has mentioned the system specific power which includes both the uranium in the reactor and outside the reactor. Now if we have an integrated plant, it contributes also to a high system specific power; so to completely rule out the integrated scheme, cuts off some of our merits of the high-temperature reactors in general.

Dr. SHEPHERD *(Dragon Project)* : May I just say, *à propos* the remark which you made a little while ago, one could conceive of having an electrolytic deconsolidation facility for separating coated particles for transportation, and making that integral with the reactor. I cannot see any reason for not thinking along those lines.

Dr. CAPRIOGLIO : That sounds very reasonable. Any further comment on this general point?

Mr. FORBES-GOWER *(CEGB)* : I wonder if I might ask some rather naive questions on this. You are speaking of having integrated fuel-handling plant with the station; it has been common with other fuels to have a decay cooling hold-up time. Is this not going to occur with these fuels? And if so, does not the delay between taking the fuel out of the reactor and one's ability to start the reprocessing, rather go against any thoughts of having an integrated plant? Secondly, if you do have to have such a delay time, how have you been costing it and who has been paying the interest charge on the capital investment of the hold-up material?

Following on from that, I wonder if I might ask the Authority a question. Recently our fuel contract has been modified so that we have negligible buy-back price for our plutonium. I was wondering what the selling price of plutonium was going to be in the future as a result of not having any price when given to them. The other thing that was puzzling me was this repeated mention of transportation and transportation costs. I am a bit naive about this. We transport all our fuel in large iron flasks on lorries; this has to go many hundreds of miles, but the cost is not so large that we do not have any operating reactor as a result of it. The fuel is all made up in the North of England and transported on a whole down to the South. Could the Authority give us perhaps some actual figure for transportation costs? Is it really the nightmare that it is being made out to be?

Dr. SHEPHERD *(Dragon Project)* : Regarding the difficulty of transportation of spent fuel from high temperature gas-cooled reactors, is perhaps based on the concept of rather large prismatic fuel elements which would be quite awkward to contain and shield. But of course we do not necessarily have to assume that this will be the case in the future high temperature gas-cooled power reactor. The fuel may be in small units that could be transported just as easily as the present fuel is shipped around from other reactor systems, in which case integrated reprocessing facilities would not be so important as in the case of large prismatic elements, 20 or 30 feet long.

Mr. WALTON *(IAEA)* : The problem of transport with high temperature gas-cooled fuel elements seems to be primarily the problem of separating the reactor moderator from reactor fuel element. It seems with high temperature gas-cooled reactors, the fuel elements often include at least a portion of the reactor moderator, and because of this the weight of fuel shipped is very low, compared to the volume of the fuel element shipment. This extra volume is required for transport of moderator. So some separation of the moderator before shipment of the fuel does seem important.

3. — HEAD END PROBLEM.

Dr. CAPRIOGLIO : If there are no more comments on this, I would suggest that we start from the beginning and take the *head end problem.* It was apparent to me in Dr. Zifferero's presentation that there are now two approaches to this head end problem, and that perhaps each of them has advantages for a particular fuel type. I have been led to understand that the grind and leach methods seem to be more appropriate if silicon carbide is used as a coating material (at least in part), while the burn and leach would perhaps have advantages for the more straight-forward pyrolytic carbon coatings. I am not sure whether this is a generalization that is possible to make. I should like to ask Dr. Zifferero if this is really the case, and to introduce a discussion of this, if need be.

Prof. ZIFFERERO *(CNEN Rome)* : Well, actually the principal option from what I gathered from the papers is between burn and leach and grind and leach methods. Actually grind and leach is a unique method to cope with silicon carbide coated particles since, in just one step, it exposes the surface to the chemical attack. Grind and leach of course could be used also for pyrolytic coated particles. But the burn and leach method can be most advantageous if the burning temperature can be reduced, and this seems to be possible according to the experiments carried out at Mol by Dr. Schmets. A particularly good combustion has been obtained there, — it is more an oxidation than a combustion — at extremely low temperature of the order of 250-300° C, by the use of a combination of nitric acid vapour and oxygen. Maybe, to start a discussion, the relative merits of burn and leach versus grind and leach should be examined first; the different options in the burn and leach methods as low temperature or nitro-combustion could be discussed later.

Mr. PODO *(Dragon Project)* : I am referring to the paper by Dr. Pierini and Dr. Schmets. I have a question. What is the general relationship between the physical state of the carbon and its reactivity with nitric acid vapours at that temperature, 320-400° C? For example, what is the relative rate of reaction between the fine grain of graphite and a dense pyrolytic carbon and a fine carbon black?

Dr. SCHMETS *(CEN Mol)* : The main purpose of the programme was not the reprocessing of carbon coated particles. We have made the experiments both on graphite and carbon material.

The data that we have given are those obtained on high density spectroscopic carbon; we did not observe significant changes in the reaction rate with graphite. No experiment has been performed on carbon coated particles as they are prepared by present technology.

Dr. CAPRIOGLIO : Do you have any further comment on this grind and leach versus burn and leach method?

Dr. ROEMBERG *(Dragon Project)* : We would be very interested to learn the advantages in the burn-leach process as compared to the grind-leach process. We have been assured that the milling equipement as specified in our conceptual design study is cheap and commercially available. Moreover, grind-leach seems to us at present the only sensible way for reprocessing particles containing silicon carbide coatings.

Mr. NICHOLSON *(Oak Ridge)* : We see the advantages of the burn and leach process as follows. First, we consider it to be a very universal process, in that it can handle the oxide kernel fuels and the carbide kernel fuels. Furthermore it could also handle a silicon carbide containing coated particles by a supplementary grinding operation. I concede that the grinding is a very simple cheap process. One point in favour of the burn leach approach is that if there is any diffusion of the uranium into the pyrolytic carbon, in such a way that it is not too accessible to the leaching reagent, the burn and leach approach will recover this uranium. Another advantage is that it permits us to discard the structural graphite as gas, thereby avoiding a long-term solid radio-active waste storage problem, because there is going to be some fission products retained in the graphite that is discharged by a grind and leach process.

One other point, we think we can get quantitative recovery of the uranium in the burn and leach approach since all the uranium is available for leaching. But in the Dragon report, it appears that the recovery of uranium is quite dependent on the fineness of the grinding operation. I would like to know, for example, how difficult really is the filtration and washing problem associated with the grind and leach approach. We have had some rather adverse experiences with that particular filtration and washing problem with another graphite fuel.

Mr. PODO *(Dragon Project)* : Well, unfortunately we have not any information on irradiated fuel, but as far as unirradiated fuel is concerned, the laboratory information is very encouraging. We have not found any difficulties during the filtration of graphite.

Mr. NICHOLSON *(Oak Ridge)* : Could you quote some filtration rates and pressure drops?

Mr. PODO *(Dragon Project)* : We generally do the leaching by using a volume to weight ratio of 2, i.e. 2 ccm of leaching solution per gram of powder. We have done experiments with 200 grams of powder where the uranium/thorium/carbon

ratio was 1/8/100. We have seen that after the second washing with water, where the volume of water used for each washing was the same as the volume of the leaching solution, practically more than 99 % of uranium was washed out.

Mr. NICHOLSON *(Oak Ridge)* : Thank you. I omitted one other advantage of the burn and leach approach. When we have done hot cell experiments with the irradiated carbides by the hydrolysis route, we have encountered severe emulsion problems due to the organic materials that have been in the solutions. Consequently, we have, on a very limited scale, used a permanganate treatment. One slight advantage of the burn leach operation is that these organic materials are eliminated in very early stages of the process and you do not have to worry about possible explosion hazards in doing a boil-down for example, to concentrate the thorium feed solutions or do a permanganate treatment to inactivate them, so that you can have a satisfactory solvent extraction operation.

Dr. CAPRIOGLIO : Is there any further comment on this one from Würenlingen or from CNEN?

Dr. SCHMETS *(CEN Mol)* : I am wondering if during leaching operation on carbides, heavy parafins are produced; and if so, has it an influence on the process?

Mr. NICHOLSON *(Oak Ridge)* : I am not sure of the identity of the material that have been produced, but I know that some wax-like materials are found in the solutions.

Prof. ZIFFERERO *(CNEN Rome)* : I have a question to Mr. Nicholson. You mentioned that you have experienced with permanganate in order to oxidize organic stuff. Could you give us some results of this treatment?

Mr. NICHOLSON *(Oak Ridge)* : I do not remember the exact details of it. It was digestion with potassium permanganate and then filtration to remove the precipitated manganese dioxide. The temperature and time and acidity are details I just do not know. But this was satisfactory in producing a feed that would operate in very small mixer-settlers without emulsification, whereas the untreated solution emulsified quite severely.

Mr. SHORT *(UKAEA Windscale)* : I think I can say something more about the products of hydrolysis which are anticipated in the Dragon process. The main residual materials in leach liquors are, from the Oak Ridge work, expected to consist of polynuclear carboxyclic acids of the mellitic acid type. The object of the permanganate treatment is to destroy about 4/5 of the residue which is left after hydrolysing and leaching, and the Oak Ridge workers found that the resulting uranium and thorium solution behaved quite normally in the solvent extraction system with tributyl phosphate.

Mr. TYTGAT *(Euratom)* : I have tried to find out from different experts on reprocessing what could be the advantages or disadvantages of a sulphochromic

dissolution of separate coated particles, because I think this solution is used to attack regularly coated particles (Florentin solution). It is done for making regular penetration of coated particles of pyrolytic carbon to find out the amount of uranium diffused in these coating. This solution has been used in Saclay and Battelle for coating studies. Would it be an impossible solution for complete dissolution of coated particles, because it works at about 80 to 90° C? There might be some problems with materials on the whole cycle, but if I could have some comments on this technique...

Dr. LOPEZ-MENCHERO *(Eurochemic)* : We have done few experiments on request for Mr. Tytgat. First of all, I would like to know if in this experiments in Saclay, they have attacked silicon coated particles?

Mr. TYTGAT *(Euratom)* : I forgot to mention, it should be only on pyrolytic carbon coated particles.

Dr. LOPEZ-MENCHERO *(Eurochemic)* : Our experience is that this solution does not attack silicon coated particles at all, but that it works with the pyrolytic carbon coated particles. When you try to keep the resulting chromic sulphate in solution, however, you have to deal with very large volumes.

In spite of its feasibility, I do not think that this type of attack can be used as a basis for head-end treatment of carbon coated particles.

Mr. TYTGAT *(Euratom)* : What are the solids you are talking about? Because from what I heard, you oxidize your carbon: so it forms CO normally.

Dr. LOPEZ-MENCHERO *(Eurochemic)* : You produce chromium (III), which either precipitates as chromic sulphate or is kept in solution by dilution. You produce about 6 kg of Cr(III) per kg of carbon.

Dr. MERZ *(KFA)* : Just a small comment to this method you just mentioned. We tried it too, and in principle it is possible, but the reaction rate is much too low to be useful for the dissolution of coated particles.

Mr. TYTGAT *(Euratom)* : My understanding is that the attack by the solution is rather rapid (a few microns per minute). I do not think that this is a slow process. In fact, I always thought it was a pretty fast dissoluting agent.

Dr. ACCARY *(Saclay)* : This method was more used to have a uniform attack on the shell, and the purpose of it was to remove concentric spheres. So the purpose is different when you try to leach out the particles. I do not think that there is a reason for using that method for leaching the particles, for removing the carbon from the kernel.

Mr. TYTGAT *(Euratom)* : No, I think the idea would be to dissolve completely the whole particles, not only the carbon but the whole fuel kernel.

Mr. NICHOLSON *(Oak Ridge)* : My personal opinion is that if we can be given the coated particles free of the structural graphite, we have two very good routes to go : this is either to burn them or to grind and leach. I would hesitate to introduce the chromic acid mixture into the mixture, because nitric acid would do just as well.

Dr. CAPRIOGLIO : When you said that there were two routes available, I was expecting you to say, to burn them or to burn them! Is here any further comment on this head end problem?

Dr. LOPEZ-MENCHERO *(Eurochemic)* : Could I ask Mr. Nicholson about any experience they have in Oak Ridge about leachibility of the uranium/zirconium oxide resulting from a burn-process for fuel containing uranium carbide/zirconium carbide?

Mr. NICHOLSON *(Oak Ridge)* : We do not have any experience with that.

Mr. MOCCIA *(CNEN Rome)* : I want to outline that the grind and leach method can be much simplified by the electrolytical deconsolidation. Although electrolytical deconsolidation seems to be an additional step to the grind and leach method, it can permit to limit the subsequent steps of the process only to the almost bare particles and to work in a wet continuous medium.

Mr. SHORT *(UKAEA Windscale)* : I would like to ask the authors of the papers on burning processes for head end, what they consider to be the problems of fission product volatilization. Both these papers state that ruthenium and caesium both volatilize within the reactor, but they deposit in the cooler parts of the off-gas system. Now what I am asking is : Wouldn't this create an intolerably high activity in the long run, by allowing the longer-lived fission products to accumulate over a long period of time ?

Mr. NICHOLSON *(Oak Ridge)* : I think, we are speculating at this stage on the exact way the fission product volatilization is going to occur. From what we have seen from the tube-furnace combustion tests with hot samples, and from out pilot plant burning of inactive graphite fuels, we conclude that the system will work in the following manner : In the settling zone above the fluid bed and before the filtration, there is a dust cloud. This is cooled and serves as an excellent collection and nucleatron agent for the condensing volatilized fission products. Consequently these things should be quite efficiently trapped by the filters. We have information that indicates that the dust-coated sintered metal filters function extremely efficiently for trapping extremely small particles. Under the reducing, high temperature conditions that exist in the upper part of the fluid bed, there is a good probability that ruthenium will not be volatilized to an appreciable extent. There may not be too much of a problem of deposition of these materials on the metal surfaces in the top of the fluid bed burner, because there is a continual reflux of dust from these surfaces back into the fluid bed. So the question really is, how high does the concen-

tration of these materials build up in the fluid bed before equilibrium is reached with the discharged amount in the product and the waste gases. We do not know what the answer to this is. We hope that the hot cell experiments that we plan within the next few months will indicate the degree of escape of the fission products to the gas space in the upper part of the fluid bed, and through the filters.

It is my personal belief, and I could be wrong, that you can contain the ruthenium and caesium and discharge them satisfactorily with the product from the bottom of the fluidized bed. But again, we need some experiments to confirm that. It is also my personal opinion that we can devise an adequate clean-up system for the off-gases that have passed through the metal filters. I cannot specify what this system is yet, because we do not know fission products the gas will contain.

Dr. SCHMETS *(CEN Mol)* : I want to make some comments about fission products escaping when using the nitric acid vapour plus oxygen process. We have obtained results indicating that about 80 %-90 % of ruthenium and 10-30 % of iodine will escape from UO_2 or carbide fuels. Now, my comment is the following : This problem of off-gas treatment for a system involving nitric acid vapour has already been solved for the existing aqueous reprocessing plants.

Dr. DE NORDWALL *(AERE Harwell)* : One quite unique feature of these fuels is the high concentration of fission products in a dissolver in a wet process, which leads me to wonder whether they will ultimately become the cheapest source of long-lived fission product isotopes such as krypton 85 and strontium 90. Krypton 85 could be very easily extracted from a grind leach process, less so from a combustion process because of the enormous volume of oxygen. The other thought that comes to mind is the ready concentration of strontium 90 in the fuel tube or matrix graphite, which again provides comparatively high concentration source of this potentially commercially valuable material.

Mr. PODO *(Dragon project)* : May I answer the question asked by Mr. Lopez-Menchero about the dissolution of zirconium oxide and uranium oxide? We have experienced in Dragon — two years ago — that when you burn zirconium and uranium monocarbide to oxide, it is possible to dissolve this product in concentrated sulphuric acid by prolonged boiling.

4. — DECONTAMINATION STEP.

Dr. CAPRIOGLIO : I suggest that we take up now the decontamination step and I feel that here we are faced first of all with two widely differing methods, the aqueous method and the dry method. I think it is very difficult to have a fruitful and long discussion on this, because the two methods are in such different stage of development, which is always making it very difficult to talk about advantages and disadvantages. But I am sure that there are things to be said, and I would like to start again from the Dragon side, see if they have any comment on the solvent

extraction step, either with respect to the aqueous versus dry method, or with respect to the complete decontamination of uranium versus partial decontamination of thorium/uranium.

Mr. SHORT *(UKAEA Windscale)* : I am not too clear how I could differentiate between the two alternate solvent extraction processes. These are reasonably well-established processes about which a good deal is known. We do not foresee any insuperable difficulties in developing these to give the required decontamination targets for the remote refabrication or for the direct refabrication. One might say that the difference between our high-decontamination system and other high-decontamination systems which have been proposed, is that we have suggested that rather than have, perhaps, two cycles of solvent extraction followed by an intermediate storage for the uranium product, and then followed by a clean-up of the uranium by an ion exchange or an absorption process prior to its use in the refabrication system, we have opted in this case for a third decontamination cycle which has a double function, insofar as it gives us the final clean-up from fission product activity and it also gives us the clean-up from thorium-228, which has bred in from decay of uranium-232 in the intermediate storage between the second and third cycles. It also has another function insofar as it can be used as a recovery cycle for wastes sent back from the refabricating lines. I think that is really all I would like to say for the moment.

Mr. NICHOLSON *(Oak Ridge)* : As far as we can tell, the aqueous processes we have devised to date should be adequate for this. From our standpoint, the major disadvantage of the volatility approach is that it does not volatilize the thorium. If you wish to have thorium recovery, then you have to go into an aqueous scheme. Then there is a question of why couple the complication of volatility on to the process to begin with.

Dr. CAPRIOGLIO : This sounds to me a very serious objection. I am wondering if there is any volatility-minded exponent here who is wishing to comment upon that.

Dr. SCHMETS *(CEN Mol)* : I agree with Oak Ridge comments about the volatility of thorium fluoride, but I think that experiments are starting in this field and it is really too early to decide something. Now I think it is absolutely not necessary to have high decontamination factors for uranium and if you want a very high decontamination, you have to pay for it. But if you want a decontamination factor in the order of magnitude of 10^4, a single uranium volatilization step will probably be enough.

Prof. ZIFFERERO *(CNEN Rome)* : As mentioned by Nicholson, previous experience in aqueous processing of thorium bearing fuels is sufficient to guarantee that this process will operate also with HTGR fuels. I am a little doubtful though if previous experience is sufficient to cope with the problem of solvent resistance

to the extremely high specific activity. I have seen that Mr. Short, in studying the reprocessing flowsheet for the Dragon Project has envisaged a rather drastic way to clean up the solvent; that is flash distillation. I should like to ask Mr. Nicholson, if he thinks that existing experience is sufficient to be safe on the solvent side.

Mr. NICHOLSON *(Oak Ridge)* : There may be a very real problem there. We have not investigated it extensively. However, in the case of solvent clean-up system, I think there is adequate technology available so that we could devise almost any system that is necessary for cleaning the solvent.

After all, one could dilute the feed, for example to give an equivalent MW day per litre as obtained in conventional power reactor fuel reprocessing. This would be one way to handle it. Another problem that has only been touched on is the fact that there probably will be insolubility problems associated with many of the fission products that are present. Some head end treatment is going to be necessary, possibly centrifugation etc. to remove these precipitated fission products. Zirconium might be one in this system, molybdenum might be another.

Mr. SHORT*(UKAEA Windscale)* : I agree with Mr. Nicholson's remarks that we do not anticipate that it would be impossible to install solvent treatments to overcome the effects of radiolysis on solvent due to the high specific activity in these sort of fuels. We have in fact done a very preliminary and provisional estimate of the damage to the solvent in the first contactor of the proposed Dragon processes, and we find from these early calculations that if this the solvent extraction is carried out in a pulsed column, the residence time is quite short and we think, at the moment, that solvent radiolysis will not be very severe.

Dr. DE NORDWALL *(AERE Harwell)* : Has any study been made on the problem of evaporating fission product solutions for storage from 150,000 MW days per ton fuel? Are there going to be limits which are going to give rise to much larger volumes than would be economically desirable ?

Mr. SHORT *(UKAEA Windscale)* : There have not been any experimental studies done on what concentration factors are achievable with the fission products from these sorts of fuels. However, at the moment we are anticipating that it will be possible to concentrate the fission product to the same sort of concentration as we achieve in the concentration of the wastes from magnox fuels. In fact, in some respects, the wastes from high temperature reactor fuels may be less difficult to concentrate because of the absence of certain salts which are present in the metal fuels. Furthermore, one of the main problems in concentrating the wastes from a magnox reactor system is the presence of residual phosphate which originates in the very large volumes of wastes which arise from a given input of fission products. Now in the case of Dragon, the volumes involved are much smaller, and hence the amount of phosphate which is present is much smaller, hence the precipitation of fission products, such as zirconium and molybdenum as phospho-molybdates, we expect, will be less severe.

Dr. DE NORDWALL *(AERE Harwell)* : May I ask, whether there is going to be a very tight limit on the quantity of thorium that you can allow to pass into the fission product phase due to the insolubility of thorium, perhaps thorium phosphate ?

Mr. SHORT *(UKAEA Windscale)* : We have not actually thought about thorium being a problem. The contacting equipment will be designed to give us a very high recovery, or at least a reasonably high recovery, and the concentration of thorium, last that we would design to, would be fairly negligible. (i.e. compared with the concentration of other metal salt already present).

5. — REFABRICATION METHODS.

Dr. CAPRIOGLIO : We can now go a step further and go to the refabrication methods. There is one thing, that I am baffled by : it is this question that was in fact raised this morning, about the many so-called sol-gel processes. We have intended this morning, to talk about *the* sol-gel process, but in fact there are various methods, which are quite different from each other, and can have various very different potentialities. I would very much like to ask the proponents of each of them to try to summarize, since he certainly knows the others, what to his mind are the best merits of the system that is being studied, by himself and by his people.

Mr. HORSLEY *(Dragon Project)* : We have, in the Dragon Project, looked at the sol-gel processes developed by CNEN Casaccia, by KEMA Arnhem as well as by Oak Ridge. We feel that each has merits which produces the best particles for, say, a different size of particle. Thus, the process developed by KEMA can be adapted to produce a 1 mm diameter thorium/uranium/dicarbide particle. The method proposed by CNEN is very convenient for consideration for use in a recycling plant where you have a combined thorium/uranium stream, because one does not have to go from a liquid phase to a solid phase and come back to a liquid phase to form the sol. For our purposes in the Dragon Project, we have combined various aspects of these laboratories' processes. Thus, for making irradiation specimens of sol-gel particles, we have of course in the Dragon Project always concentrated on trying to produce porous particles where most other laboratories always try to produce dense particles. With this in mind, we began by making sols with a method that has been outlined in our report, which has been developed from the method that has been used at KEMA, and we are able to gel these sols by a number of methods. We have used both the internal gelation method and the dehydration method. Either of these methods has advantages, depending on what size of kernel you wish to make. If you want to make a very large one, personally I would prefer an internal gelation method, whereas if I wanted to make a very small one, I would prefer a dehydration method. And so the route that you choose depends very much upon what kind of kernel you want to have.

This brings me to a major point which I hope will be further discussed. This is, just what do people reckon is the optimum size for a fuel kernel? Because it is our experience that the choice of a fabrication route may be determined by the size of particle we wish to have. If we want a large one, we find the powder metallurgical method most efficient. If we wanted a very small one, maybe we would go to the sol-gel. Coming back to the sol-gel process itself, we have produced both porous carbide and porous oxide kernels and I would like to suggest, so far as refabrication is concerned, that the way one refabricates may have an influence on the way one does a further recycle, because we have heard this afternoon of the difficulty of dissolving up thoria/urania dense microspheres. If we make it porous to begin with, maybe there is a good chance of getting over this dissolution problem. Porous microsphere of thorium/uranium can be successfully made up to quite large diameters.

Dr. CAPRIOGLIO : This problem of kernel size, which we have skipped this morning, I think it is most properly going to be discussed now. May I ask the CNEN people if they wish to add anything to what Mr. Horsley just told us ?

Mr. COGLIATI (*CNEN Rome)* : From the point of view of gelation methods, I would like to note that we are able to prepare spherical particles of uranium dioxide by extraction of nitric acid from the droplets of the sol. The sol is dispersed in the organic medium containing some liquid ion exchanger. In this manner it is very easily possible to obtain for the uranium dioxide particles with a diameter up to 500 microns. The same method can be used also for uranium and thorium mixed oxides or carbides. Uranium and thorium can be in any ratio, because our method is based on the preparation of a sol that contains uranium in the 4th state, not in the 6th state. In this manner, no problem arises from the heat treatments and for the reduction of uranium in the furnace by hydrogen.

Mr. VAN DER PLAS *(Kema)* : I feel that the situation as to the various processes now available, has been well outlined by Mr. Horsley. It is very much my feeling that it depends really on the customer's total fuel cycle, whether he wants to incorporate a sol-gel method at all in his fuel cycle and which of the various variants now available he wants to choose. I would like only to point out one disadvantage that is common to all variants now known. That is the treatment of the aqueous waste and the recycle of the dispersion liquid. I have no experience on that; we are not really in for production, only for research, but I know that this gives problems and is perhaps a detail worthwhile to consider for a moment.

Mr. DOUGLAS *(Oak Ridge)* : I also would comment that the variations in the sol-gel process have already been very nicely outlined. The schematic of the particular process that was conceived by Dean and Ferguson at Oak Ridge is shown on Page 172 of the paper by Lotts and myself, and I do not see any point in discussing or describing it. It does have certain advantages in that the sols that are produced are very stable. The absence of the internal gelation means that there is no time problem

as far as the production of microspheres is concerned. Likewise we do not have a washing step to go through, there are no problems of drying of the oxide microspheres as produced. Our objective has been to make very dense particles. The reason for this is that we find a very keen relation between the cost of fabrication and the number of individual units that have to be processed. So, therefore, with a 100 % dense particle you obviously have to coat fewer particles for a given charge in a reactor. The solvent that we use in our dehydration process is alcohol: therefore, it is very cheap. On our recycling process we have been able, by use of a very simple distillation step, to recycle the solvent and extract the water and maintain it at an acceptable level in the solvent. This in our mind does not pose a difficult problem. Concerning the size of the particular particles, as I stated this morning, we completely agree that if your desire is to make very large particles, then either internal gelation is required or else a powder metallurgy process. In our own mind there is some question about the desirability to go into larger particles. A somewhat abstract analysis has indicated that as one goes to larger particles, there is an increase in the cost of coating them. Therefore, there is some penalty attached to the selection of a larger size particle. We also are not quite sure what the performance will be of larger particle sizes. It may be that a heavier coating is required if one only uses pyro-carbon. Perhaps this is one advantage with silicon carbide. Due to its higher strength perhaps no additional thickness of coating would be required.

6. — KERNEL AND PARTICLE SIZES.

Dr. CAPRIOGLIO : I was very interested by this comment about the optimum size that seems to be crystallizing in the minds of the Oak Ridge people for the kernels. I would like to ask now which is, to their mind, this optimum size; there is obviously an incentive to have large sizes in general terms and since there is apparently a drawback in having very large sizes, may I ask what sizes are we talking about ?

Mr. LOTTS *(Oak Ridge)* : I might say that the analysis of this was quite theoretical, but we think that on the order of 300 microns would be the size that would minimize the coating costs. I might just indicate what is already indicated in the report, to emphasize the basis for the analysis. First of all, it is true that the amount of coating per kg. of particle is less if the coating thickness is the same as you increase the particle size. But, along with the increase in particle size you have increased amount of methane, which you discharge into the furnace. Therefore our conclusion from this is that a coater would have less capacity with large sizes.

Mr. HUDDLE *(Dragon Project)* : The whole question of particle size is, I think, one where it is not yet possible to given an answer from experience; it can only be speculation. In our programme, we are trying to cover the whole size range, since our objective is to provide the Signatories of the Dragon Agreement with those facts that are important to a high temperature reactor system. As yet, we

have seen no significant effect of size in our irradiation programme. We have, however, noticed an effect concerning shape: this is one of the major factors in favour of the sol-gel process, since it produces excellent spheres. Coming back to size, I think this will depend very much on the design of the actual fuel, and will be associated with the fuel loading required in the compact. In Dragon, we favour larger particles and thicker coatings, and we would suggest a particle having a kernel of 500 to 600 microns in diameter with a coating very much thicker than is presently suggested. I would go for a complex coating of about 150 to 250 microns in thickness.

Regarding the question of fluidized beds and the problem of sooting, it is my opinion that, these are purely transitory phenomena, since by the use of appropriate sooting inhibitors the problem can be eliminated. Incidentally, the use of certain of these inhibitors leads to improved rates of deposition, thus increasing markedly the capacity of existing equipment.

Mr. SHENNAN *(UKAEA Springfield)* : I would like to comment on the effect of kernel size on coating efficiency on the basis of our experience at Springfields which contrasts with the results of the Oak Ridge theoretical analysis. We have examined pyrocarbon deposition on kernel sizes ranging from 230 to 650 microns. Although there is a large variation in the specific surface area, the efficiency of pyrocarbon deposition in a fluidized bed remains constant for reasonably large bed volumes. This result is easily understood when it is assumed that the deposition zone is limited to the region close to the gas inlet, coating being a cyclic process, for comparatively large bed volumes, the particles moving continually in and out of the reaction zone. Thus, a lower specific surface area simply reduces the specific deposition rate, i.e. increases the volume of the reaction zone. Provided this reaction volume is less than the volume of the charge, no change in efficiency will be observed. However, the increase in reaction volumes obtained with large kernel sizes does result in small changes in the structure of pyrocarbon coating.

Mr. JAYE *(General Atomic)* : I would like to ask Mr. Huddle one question. He mentions that he finds no effect of particle size as to stability of the particle, I believe. Does he keep the coating thickness in proportion to the particle diameter, or does he let it get smaller, relative to the particle diameter?

Mr. HUDDLE *(Dragon Project)* : I do not think that the accuracy of the results in such irradiation tests warrants a conclusion on this matter. I was implying that in our tests small particles of about 200 microns had behaved in a similar manner to those of about 600 microns. At least, we noticed no significant effects. I cannot answer quantitatively but I think it would be very difficult and, at this stage, even dangerous, to draw any major conclusions. At present, it is a question of collecting operational data and this we hope to do in the course of our programme in the Dragon Reactor Experiment.

Mr. JAYE *(General Atomic)* : I would just like to point out the importance of this variable of the thickness of the coating to size of the substrate. I might say

as a reactor designer, I am interested in getting fuel into the reator, and since this particular variable goes as a cube, as the thickness of the coating can be reduced relative to the thickness of the substrate, we can make very rapid gains indeed, particularly for practical long exposures with fuel zoning where some very heavy loadings are required at certain locations.

Dr. SHEPHERD *(Dragon Project)* : This was in fact one reason why we went to larger particles; it was hoped to have larger particles with the same thickness of coating in order to get more metal into the compacts.

Dr. EATHERLY *(Union Carbide)* : I might recount a little of our experience with going to larger and larger particle sizes. In particular with the dense fused core particles that we have used, we run into a thermal shock problem at about 4 to 500 microns in diameter. The problem becomes so severe above 500 microns that the particles split immediately upon entering into the furnace. So this is certainly one disadvantage of large diameter, very high density cores. The second problem that we have run into is that as we scale up the furnace size, it becomes harder and harder to avoid the sooting problem for the large diameter particles. We have no difficulty at the 2 inch furnace level, coating particles around 500 microns. In the 5 inch furnaces, our coating structures change very, very radically, and we have found it almost impossible to eliminate the sooting.

7. — REMOTE REFABRICATION.

Dr. STOECKER *(KFA)* : I think that reprocessing specialized on one fuel type has a distinct advantage, and this seems to be especially true for the pebble bed reactor, because it has the simplest fuel element, especially in connection with the use of loose particles. We even envisage the re-use of graphite balls, and are developing a machine which can take out burnt loose coated particles and fill refabricated particles in the ball. Therefore, we are interested in techniques for complete reprocessing and remote refabrication of coated particles. I would like to hear any comments from the reprocessing people if this seems possible at a reasonable price.

Mr. NICHOLSON *(Oak Ridge)* : I do not believe from the reprocessor's standpoint that there is any essential difference in the processing steps required for either the prismatic fuel or the ball-type fuel. They are both reducible to the same processing system, if you mechanically dismantle them and remove the particles. It is only a matter of size of the equipment to remove the particles from the respective fuel elements. As far as the cost is concerned of reprocessing, our study indicates the cost penalty that is attached in putting a head end on to an existing process building which was designed for another type of fuel. A large process plant, as we indicated in our study, that is specifically designed for HTGR type fuels, should not cost much more than the present NFS system.

Mr. DOUGLAS *(Oak Ridge)* : For the refabrication end of the fuel cycle line, I would comment that there is need for some consistency. I would plead with the

reactor designers to try to achieve some standard; not necessarily in the design of their reactors or even in the shape of their fuel elements, but at least in the way that they will accept compacts of coated particles into their fuel geometrics. There is no reason why a wide variety of fuel elements cannot be refabricated because all of the steps until the final inclusion of the coated particles in the graphite matrix can be achieved using equipment in common. Particularly the sol-gel process is very flexible, both in terms of making different sizes of spheres and in changing composition. Likewise the fluidized-bed coater can easily accomodate variations in coating requirements; thus, a variety of different fuel concepts can be made. But, I would caution that it would be very difficult to make a common facility in which the final assembly process was vastly different. For instance, present ways of making molded fuel balls for AVR versus the compaction techniques for Dragon, are so different that you would have difficulty in having an economic refabrication plant. But I see no reason why they could not have a similar enough design so that a refabrication plant could serve both concepts.

Dr. SHEPHERD *(Dragon Project)* : It is not my intention to be drawn into an argument about the respective merits of prismatic versus spherical fuel, but I would remark that the cartridges in Dragon and in Peach Bottom are roughly the same size as the balls of the AVR reactor; so one is concerned only with a matter of difference in shape. I cannot see that the problems are going to be very much different in fabricating either.

Dr. SCHMETS *(CEN Mol)* : I think that two steps of dismantling the fuel and fabricating a new fuel are steps where an important part of the manpower of the site is included. My feeling about this problem is that, integrated plants including reprocessing and refabrication, would be more economical. The transportation costs to and from the reprocessing plant would be canceled and the reprocessing tail end could be combined with fuel fabrication.

8. — LOOSE PARTICLES VERSUS COMPACTS.

Dr. DE NORDWALL *(AERE Harwell)* : I would like to come back to the point which was made a little earlier about getting the fuel into the fuel element. First of all, I would like to ask whether the problem exists at all with loose fuel where very much higher ratings are possible, due to better packing and the lack of necessity for leaving space between the particles. And secondly, to make a comment, that is that the potential use of UO_2 in the kernel at rather higher operating temperatures than are envisaged for carbide fuels, would allow thicker compacts with fuel kernels of the same diameter.

Prof. SCHULTEN *(KFA/THTR)* : I believe that a limit for the specific power, that means, for kilowatts per gram, is essentially due to the protactinium poisoning. The thermal conductivity of the loose coated particles is high enough to give a very high specific power. We have now only experimental data for heat conducti-

vity up to 700° C, but when we extrapolate, then we can see that there are no really serious problems as far as heat conductivity of loose coated particles is concerned. The problem is the protactinium poisoning which becomes high, when the specific power is higher than, say, 3 kW per gram.

Dr. CAPRIOGLIO : Any comments from Oak Ridge on this point of loose particles versus compacts and specific power?

Mr. DOUGLAS *(Oak Ridge)* : I have no technical comment because our concern with this concept has not been, "Is it a desirable one?". We think it is, but we wonder what will be the attitude of the reactor operator. What happens if, by some odd chance, one of these graphite structures breaks and we spill loose coated particles in the reactor? The time involved in trying to clean up the system may far off-set the advantage of having loose coated particles even if there is only one occurrence in the total lifetime of the reactor.

Prof. SCHULTEN *(KFA/THTR)* : It is necessary to investigate this question very carefully. We expect, after our first experiences, that the number of broken balls will be very, very small. In the case where one ball breaks, then we have about one gram of uranium-235 in the reactor, and about 10 grams thorium, and this is from the standpoint of material in the reactor, or from the standpoint of activity not too much. I cannot really believe that it is a safety problem, a problem, let's say, of criticality. But it must be investigated in connection with the blowers and with the whole circuit.

Dr. CAPRIOGLIO : I do not think anybody was thinking of criticality problems. It is just that people do not like coated particles going around in a primary circuit. I would like to come back to reprocessing at this point and ask, if there is any question to be raised on the reprocessing papers that have been presented today, before we adjourn for today.

9. — DECAY TIME AND BURNT-OUT FUEL.

Mr. FORBES-GOWER *(CEGB)* : I wonder if I could re-ask my question to which I received no answer. I wonder if perhaps, to crispen the answer, I could ask Mr. Douglas and Mr. Lotts directly, concerning the effects of delays between fuel coming out of the reactor and going into the reprocessing. I must apologize for not having read their paper very thoroughly, which I received this morning, but I see they do allow a 90 days delay on page 189, but I do not find any cost item for it any-where. Is this delay affected by the type of fuel that was in use or is it purely a radio-active decay delay before you can handle it, or does it allow anything for cooling? Because if we were to transport the fuel, we could not transport it while it was still hot, in the thermal sense. We do not make great allowances at present for cooling fuel during transport, due to the difficulty of assuring that it is guaranteed in the event of an accident during transport. As any of us who have worked on fuel cycles are

aware, the interest charges on fuel are very high, and can easily equal or be greater than the fabrication costs and the refabrication costs. I should like to know how much this does affect the refabrication costs, since it must be very large component of it, even for 90 days. If on the other hand one gets round the issue, by having zero price for the spent fuel, has this been taken into account in the fuel cycle cost calculations that have been done by the designing side, or the physics assessment side?

Mr. NICHOLSON *(Oak Ridge)* : I do not want to attempt to say anything about the cost aspects, but from the reprocessing and shipping standpoint, the decay is desirable, because it lets some of the very high energy gamma emitters, such as those from the barium-lanthanum chain decay away, and then in the reprocessing plant itself, it is usually desirable to have perhaps 90 days cooling to control emission in the reprocessing plant. This iodine emission will occur with either of the main proposed leaching processes and be quite a problem in the control of the plant.

Mr. LOTTS *(Oak Ridge)* : I would like to point out that we did not include any charges for interest for fuel in our paper. These are taken into account in our studies at Oak Ridge, usually in the computation of the total fuel cycle cost. I think, Carlsmith will probably speak on this tomorrow. The other thing I would like to point out is that this was a design basis for the TURF facility, the reason being that we would have to get our fuel from some reactor at the site which is distant from Oak Ridge.

Mr. ROSENTHAL *(Oak Ridge)* : The reason why Mr. Lotts omits his costs as he said is that we end up putting them, assigning the out-of-power inventory cost to the reactor operators. It seems like a reasonable assumption that since it will continue to be owned by the operator, the penalty for its inventory should be assigned to him. I just might give you an estimate of what it might cost you in one particular case for an HTGR :

The out-of-power inventory for one year is a penalty of about 0.07 mills, and using a large industry such as Mr. Lotts used, the fabrication costs is 0.26 mills. So, indeed, it is equal to about one fourth of the fabrication cost. In doing these things, what we normally do is to optimize the shipping cost by designing a cask, and then varying the design so as to determine what the minimum is for the fueling time. And when we do this, we get numbers like the 90 to 120 days, which are acceptable from the viewpoint of shipping, but also approximately minimum on a basis of the cost of building and owning casks.

Dr. WIRTHS *(Nukem)* : We are quite concerned about interest on capital. I wonder why this point has been made here for burnt-out-fuels. The value of burnt-out-fuels just taken out of the reactor is almost zero.

Dr. CAPRIOGLIO : Well, I wonder if anybody wants to comment about that. Perhaps the typical answer would be : "It depends".

Mr. JAYE *(General Atomic)* : I hope Dr. Wirths is wrong. First, if the value of the burnt out fuel were almost zero, we would not bother to reprocess it. Now, we are all working very hard to make very high converters, and we even talk occasionally about breeders, which means that the value of the burnt out fuels is indeed very high, and therefore I think it is important that this reprocessing and shipping time be optimized as Dr. Rosenthal of Oak Ridge pointed out. It was rather interesting to me that the optimization was done on a shipping cask and did not include the degradation of the solvent in the chemical plant.

Dr. WIRTHS *(Nukem)* : I can only tell by our experience; what I have said about the value of burnt-out-fuel pertains to MTR fuel, which has been burnt out at 30 %. That is really almost zero.

Mr. SHORT *(UKAEA Windscale)* : I would also like to add the comment that an optimization which does not include the reprocessing is liable to overlook problems; even at 90 days' cooling, iodine could be a very serious problem in a reprocessing plant.

Furthermore, when the fuel comes out of an HTGR, about 10 % of the uranium-233 is there as protactinium, and a long cooling time is necessary to recover a high percentage of the uranium as well. I have not done a calculation on what the recovery would be at 90 days, but I suspect it would possibly be quite a sizeable loss.

URANIUM-235/THORIUM FUEL CYCLES IN GRAPHITE MODERATED SYSTEMS (1)

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ABSTRACT

This paper presents results from calculations on both once-through and reprocessing fuel cycles. Different fuel management schemes have been considered. Fuel costs are evaluated for a wide range of values for the cost parameters involved.

1. — INTRODUCTION

One of the most important features concerning the fuel cycle in high temperature graphite moderated reactors is the use of coated particle fuel which tends to give almost homogeneous fuel elements from a nuclear point of view. In this system it is no longer possible to reduce the effective resonance integral of U^{238} by lumping to the extent that natural or low enriched uranium can be used successfully as fuel. It appears that enrichments in the order of 7-10 % are required in order to reach acceptable burn-up figures.

The attractiveness of the thorium fuel cycle in this system derives from the facts that :

(i) Once uranium has been enriched to 7-10 % the additional costs of enriching to 93 % are comparatively low.

(ii) Replacing U^{238} as fertile material with thorium makes a better fuel cycle performance possible. The thermal α-value (capture-to-fission ratio) of U^{233} is ≈ 0.10 independent of moderator temperature, whereas it is about 0.5 for Pu^{239} at moderator temperatures above 1,000° K (compared to 0.37 in water reactors). The corresponding η-values are 2.28 and 1.9. The superior characteristics of U^{233} as fuel compared with Pu^{239} are the basis for the interest in the thorium cycle.

The advantage of the high temperature reactor system is its high thermal efficiency together with the ability of the fuel to allow high burn-up (inMWd/tonne) at a high fuel rating. This paper is intended to illustrate the burn-up performance of different fuel cycle arrangements in an HTR and to show what bearing the core design parameters and the economic considerations have on the fuel costs.

(1) Dragon Project Report 336, April 1965.

2. — The neutron balance in an HTR on the U^{235}/Thorium cycle

In order to make the thorium fuel cycle economically attractive it is desirable to utilise the fertile properties of thorium as much as possible. The main factors influencing the conversion are discussed below.

2.1. — The $\bar{\eta}$-Value.

The capture-to-fission ratios (or α-values) in both U^{233} and U^{235} are higher at epithermal energies than at thermal. Consequently, the overall α-values are lower the smaller the epithermal-to-thermal flux ratio is. In other words, the better moderated a system is the more fissions are produced when a given quantity of uranium is burnt.

At the same time the η-values increase and make more neutrons available for conversion. The following Table gives η-values for the two isotopes for some carbon-to-uranium ratios (S-values) :

S		2,000	5,000	10,000	15,000
η	U^{235}	1.83	1.94	1.98	2.00
	U^{233}	2.19	2.22	2.24	2.25

Thus the overall η-value in a system

$$\bar{\eta} = \frac{\nu_{U233}\,\bar{N}_{U233}\,\sigma_{f_{U^{233}}} + \nu_{U235}\,\bar{N}_{U235}\,\sigma_{f_{U^{235}}}}{\bar{N}_{U233}\,\sigma_{a_{U^{233}}} + \bar{N}_{U235}\,\sigma_{a_{U^{235}}}}$$

increases with the S-value and the $U^{233} : U^{235}$ ratio.

2.2. — Non-Fertile Neutron Losses

(i) The probability of neutron capture in the graphite increases with higher carbon-to-uranium ratios. At S-values around 7,000 there is a break even between the increase in η-value and the corresponding neutron losses in the moderator.

(ii) Neutron losses through leakage depend mostly on the core and reflector arrangements but increase somewhat with S-value because of higher thermal leakage.

(iii) Neutron losses in the slowly saturating fission products increase with burn-up as these isotopes accumulate in the system. At thermal energies these losses compete on equal terms with absorptions in the fissile isotopes. However, epithermal neutron captures in fission product resonances become more pronounced going towards undermoderated systems.

(iv) During the running of the Reactor Xe^{135} is produced by fission and Pa^{233} from neutron captures in thorium. These isotopes decay, and the lower the flux is at the energies where their respective cross sections are high, the larger is the probability that they will decay without having caused parasitic neutron losses.

Xe^{135} is a thermal absorber. Since the thermal flux in a system is proportional to the fuel rating it follows that neutron losses in this isotope increase with higher ratings.

Pa^{233} captures also some neutrons at thermal energies but the majority of neutron losses in this isotope occur in the resonance region. Hence the protactinium poisoning of a reactor depends mainly on the power density, since the epithermal flux is proportional to this quantity. It should be stressed that the loss of one neutron in protactinium means two neutrons lost in conversion, since all neutron captures in thorium that produce subsequently burnt protactinium must be regarded as non-fertile.

(v) Finally, neutrons are lost in non-fissile and non-fertile heavy isotopes that build up during burn-up, such as U^{236}, Np^{237} and Am^{243}. Within each cycle these isotopes behave as fission products. Fuel reprocessing will separate all these poisons from the fissile uranium except the U^{236}.

2.3. — THE CONVERSION RATIO.

Each fissile absorption will produce ($\bar{\eta}$-1) excess neutrons. These will be distributed according to

l_{gr} graphite losses (increase with S-value)

$l_{Xe\,135}$ Xe^{135} losses (increase with fuel rating)

$l_{Pa\,233}$ Pa^{233} losses (increase with power density)

l_{fp} fission product losses (disregarding Xe^{135}, increase with burn-up, decrease slightly with increasing S-value)

$l_{U23^{\cdot}}$ U^{236} losses (depend on burn-up and recycling scheme)

l_{leak} leakage losses (decrease with increasing reactor size, increase slightly with S-value).

$l_{control}$ control lossses (depend on the fuel management)

The neutrons available for conversion are obtained as :

$$C = \bar{\eta} - 1.0 - (l_{gr} + l_{Xe135} + 2 \times l_{Pa233} + l_{fp} + l_{U236} + l_{leak} + l_{control})$$

and can be used for fertile captures. These mainly occur in thorium, but to some extent also in U^{234}, U^{238} and Pu^{240}.

Neutron captures in thorium lead to the formation of Pa^{233} which decays into U^{233} with a half-life of 27.4 days. Because of this time delay, fertile captures in thorium that occur towards end of life of a fuel element will not result in actual production of U^{233} until the fuel element has been discharged from the Reactor.

Defining the "in-core conversion ratio" as :

$$c = \frac{\text{Production}}{\text{Destruction}} \text{ of fissile material in the core}$$

we have that

$$c = C - l_{disch}$$

where l_{disch} expresses the loss in conversion due to protactinium discharge from the system. This loss decreases with longer fuel residence times.

For once-through cycles the in-core conversion ratio is the important one. However, in reprocessing cycles all the converted U^{233} will be recovered regardless of whether it was produced in the core or after fuel element discharge. Consequently, on this cycle all excess neutrons can be effectively used for conversion.

Conversion ratio optimisation implies finding a fuel cycle arrangement which gives a high $\bar{\eta}$-value in combination with low neutron losses and this again must be combined with a high burn-up for economic reasons.

3. — General Assumptions in the Fuel Cycle Calculations

3.1. — Core Data.

All our burn-up calculations refer to a core with the following dimensions :

Core height	4.14 m
Core diameter	6.20 m
Reflector thickness	0.80 m

These figures were arrived at in an engineering design study [1] on a station with a total thermal power of 1,250 MW and a core power density of 10 MW/m³.

The core dimensions determine essentially the neutron leakage which in our case is around 3 %. Naturally our fuel cycle results apply to other systems with different dimensions provided that the neutron leakage is the same.

The following assumptions were made in all calculations :

Fuel temperature	1,500° K
Moderator temperature	1,300° K
Graphite atomic density	6.7×10^{22} atoms/cm³
Graphite absorption cross section at 2,200 m/s	3.75 mb
Buckling	5.3×10^{-5} cm^{-2}
Fuel element geometry	completely homogeneous

The sensitivity of our results to changes in these assumptions is discussed in Section 6.

3.2. — Cost Analysis Assumptions.

The following standard values have been used for the evaluation of fuel costs :

Uranium price (93 % enriched)	£5,000/kg fissile
Thorium price	£ 15/kg
Purchase and fabrication of graphite parts	£ 1/kg

We have assumed costs for fabrication and reprocessing of fuel to depend only on the quantity of heavy metal (uranium and thorium) to be handled and not on the composition of the fuel itself. Cost analyses have been made over a fairly wide range of values for these items. We have generally kept the interest rate around 6 % and 7 % but investigated the effects of lower and higher interest on the results. Compound interest is used throughout.

3.3. — Methods of Calculations.

All calculations assume a point burn-up model which is adequate for survey assessments of this kind. Cases with discontinuous fuel handling were calculated with Helios [2] (fuel cycle assessment) and Electra [3] (cost analysis). The continuous charge/discharge analyses were made with Bass [4].

The nuclear data on the Dragon Library II [5] tape were used throughout with explicit treatment of the 28 most important fission products. No noble gas release was assumed.

It must be underlined that all our results refer to zero-dimensional calculations. For this reason axial and radial flux variations are not included in power peaks quoted here.

3.4. — Fuel Cycles Considered.

Since we do not know when reprocessing facilities will be available for HTR fuel our investigations have had to cover both once-through cycles and fuel reprocessing cycles. Our main efforts have so far been concentrated on homogeneous fuel but we have also looked into the possibility of improving the burn-up performance by keeping fertile and fissile materials separate in the core.

The following cycles have been considered :

(*a*) *Once-Through Cycles* :
- (i) Homogeneous fuel.
- (ii) Seed blanket arrangements.

(*b*) *Fuel Reprocessing* :
- (i) Homogeneous fuel.
- (ii) Segregation between fissile and fertile isotopes.

4. — Once-Through Cycles

These calculations assume that no reprocessing facilities are available and no credit is given for spent fuel. With this philosophy it is necessary to get as much power as possible from a given fuel investment and, since the initial capital outlay mainly lies in the cost of U^{235}, this implies that low fuel costs are always associated with high fifa burn-up (fissions per initially invested fissile atoms). Conversion is only of interest as a means of achieving this.

4.1. — Homogeneous Fuel.

We have considered fuel handling arrangements from complete core replacement over zoned systems to continuous charge/discharge over a wide range of power densities and fuel ratings. The fuel itself consists of a homogeneous mixture of thorium and uranium.

4.1.1. — *Complete Core Replacement.*

The whole core is charged at the same time and run at full power until k_{eff} drops to 1.0. The used fuel is then removed and a complete new charge loaded into the system.

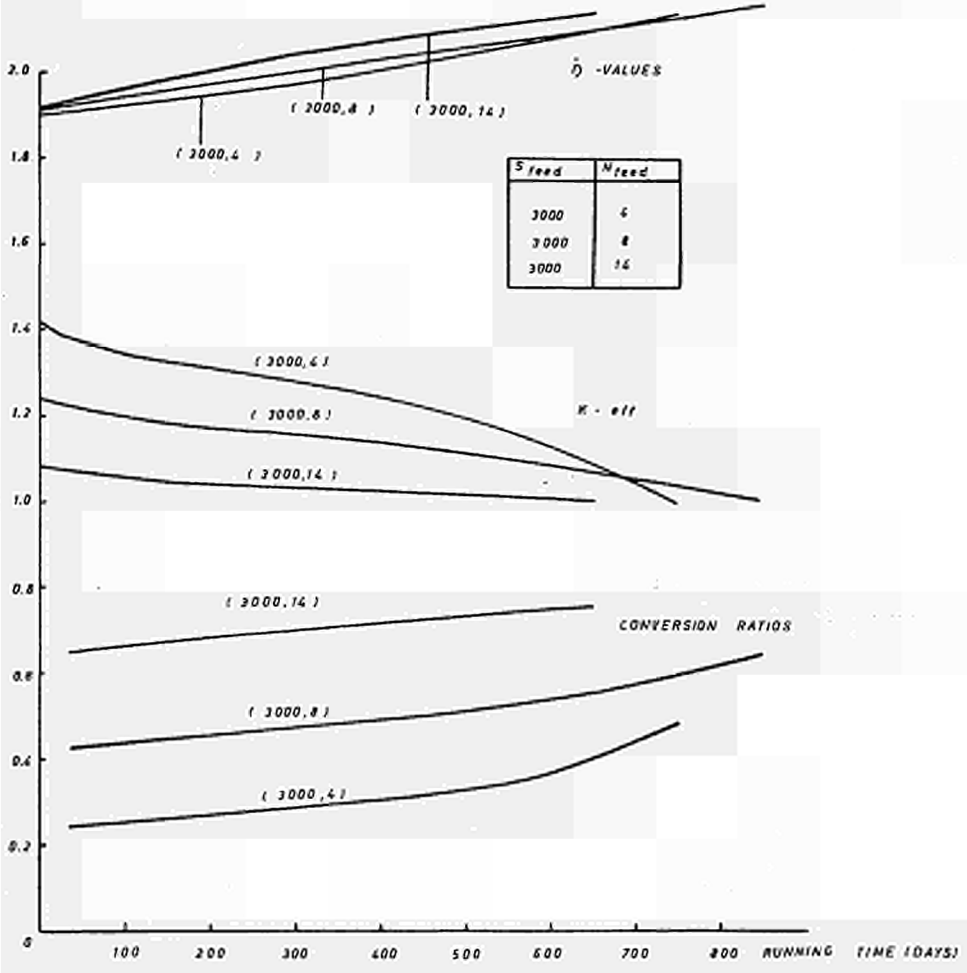


FIG. 1. — Complete core replacement. K-eff, $\bar{\eta}$ and conversion ratio during core life for some feed compositions, 10 MW/m³. Once through cycles.

(*a*) Conversion.

At any time during core life the conversion ratio is :

$$\frac{\text{Rate of Production of fissile material}}{\text{Rate of Destruction of fissile material}}$$

In this expression the denominator is essentially constant because the power level is the same.

Fissile material is produced by neutron captures in thorium both at epithermal and thermal energies. The epithermal production rate is proportional to the fast flux and thus essentially constant (the fast flux depends on the power density which does not change) whereas the thermal production rate increases with burn-up (the thermal flux increases with depletion of fissile material in order to maintain constant power). Hence the overall conversion ratio increases all the time with burn-up.

(*b*) Reactivity.

Conversion ratio and reactivity are complimentary quantities in the sense that an increase in conversion will cause a corresponding reactivity drop. Thus reactivity is lost on this cycle not only by the build-up of fission product poisoning but also through the increase in conversion ratio. Further, if one wants to give the system a high initial reactivity to obtain high burn-up, this implies a corresponding loss in the initial conversion ratio.

It follows from (*a*) that the less conversion that occurs thermally, the slower will the conversion ratio increase with burn-up and hence the longer will the system remain critical. This is an argument for putting as much conversion as possible into the epithermal region by going towards undermoderated systems. (The cycle would be ideal if all conversion occurred epithermally.). However below S-values of 2,000 the η-values start to drop and hence overall reactivity will be lost.

The fairly high initial reactivities required to obtain reasonable burn-up implies that a large amount of neutron losses occur in control poison.

Fig. 1 shows k_{eff}, and conversion ratio versus time for a few cases with S_{feed} = 3,000, 10 MW/m^3 and different feed N-values. We see that a high N-value gives a high initial conversion ratio, low excess reactivity and a correspondingly lower running time. Decreasing the N-value will increase the excess reactivity and the running time at the expense of the conversion ratio until the latter quantity becomes so low that the running times decrease again for lack of converted U^{233}.

It is interesting to note that systems with larger N-values allow a high conversion ratio at end of life. The η-values being roughly the same in the different systems, this shows how much higher the fission product losses are in low N-value cases due to the stronger uranium depletion.

(*c*) Results.

We have made calculations on this system for power densities of 5 MW/m^3 and 10 MW/m^3 over a wide range of initial N- and S-values.

The following optimum cases were found :

Power density, MW/m^3	5	10
S_{feed}	3,000	3,000
N_{feed}	8	8
Running time, days	1,844	850
Fifa	1.21	1.11
Fuel cost, d/kWh	0.193	0.190

The fuel costs were calculated assuming a fabrication charge of 150 £/kg heavy metal, 80 % load factor and 6 % interest on capital investment.

The higher fifa obtained for the lower power density is entirely due to the increase in conversion because of the reduced xenon and protactinium poisoning. However, the improvement in fuel cycle performance does not reflect in fuel costs because of the higher interest charges over the longer running time.

(*d*) Conclusions.

This system requires a high initial reactivity and has an inherent bad neutron economy because of large parasitic losses in control poison. The fuel costs must be considered high.

4.1.2. — *Zone Reloading of the Core.*

The systems considered here assume that instead of replacing the whole core when k_{eff} has dropped to 1.0 only a part of it is reloaded.

4.1.2.1. — Zones and Cycle Time — The Equilibrium Cycle.

The core is divided into M zones. At each reloading occasion the particular zone containing the fuel elements that have stayed the longest time in the Reactor is reloaded. The reactivity thus added to the system will allow it to run until the next reloading occasion. Denoting the overall residence time of a fuel element in the system T (years) we obtain the cycle time between two successive reloadings :

$$T_{cycles}\,(\text{years}) = T/M$$

The expression "zones" must not be taken literally. It refers to a group of fuel elements that are loaded and discharged together in positions scattered over the whole core. In this respect M defines the ratio between the total number of fuel elements in the core to those reloaded each time.

All following considerations refer to "equilibrium conditions" which have developed after each zone has been recharged two or three times. Optimisation of the "running in" period is a problem that has to be tackled separately once a suitable equilibrium cycle has been found.

4.1.2.2. — Reactivity and Neutron Balance.

The larger the number of zones in a system with a given fuel rating, the shorter becomes the cycle time and the less excess reactivity has to be added to the system at each reloading. In the limit when $M \to \infty$ (continuous charge/discharge) both the cycle time and the reactivity swing go to zero.

The smaller the reactivity swing, the less neutrons will be lost in control absorbers, hence the neutron balance improves with increased number of zones.

4.1.2.3. — Neutron Spectrum and Flux — The Irradiation History of a Fuel Element.

In these systems fuel elements of all ages will be so intimately mixed that the flux and the neutron spectrum do not longer depend explicitly on the composition of the feed material but instead of that of the equilibrium core. From this follows :

(*a*) The power density of a fuel element is essentially proportional to its fissile content (because the fission cross sections for U^{235} and U^{233} are about the same). Hence the rating is the same in all the fuel elements.

(*b*) The production rate of U^{233} is constant over the lifetime of a fuel element (neglecting thorium depletion and the protactinium transient at beginning of life).

4.1.2.4. — Fifa Burn-Up and Power Peaks in Fresh Fuel Elements.

The conversion ratio in a fuel element is unity when U^{233} has reached equilibrium concentration (production rate = destruction rate). Such a fuel element is in itself subcritical and a sufficient amount of U^{235} has to be provided in the feed material to keep the system at criticality.

The higher burn-up that one wants to achieve in a fuel element, the longer one would like to leave it in the subcritical state in the Reactor (to extract more power from it) and, consequently, the longer will it have to be fed with neutrons from the fresh fuel elements. This can only be achieved by adding more U^{235} to the feed. However, since :

$$\text{fifa} = \text{const.} \times \frac{\text{fuel element lifetime}}{\text{fissile investment in feed}}$$

it pays to do so as long as an increase in U^{235} investment correspond to a proportionally larger increase in running time. However, the more U^{235} that the feed material contains, the higher will the initial power density be in these elements.

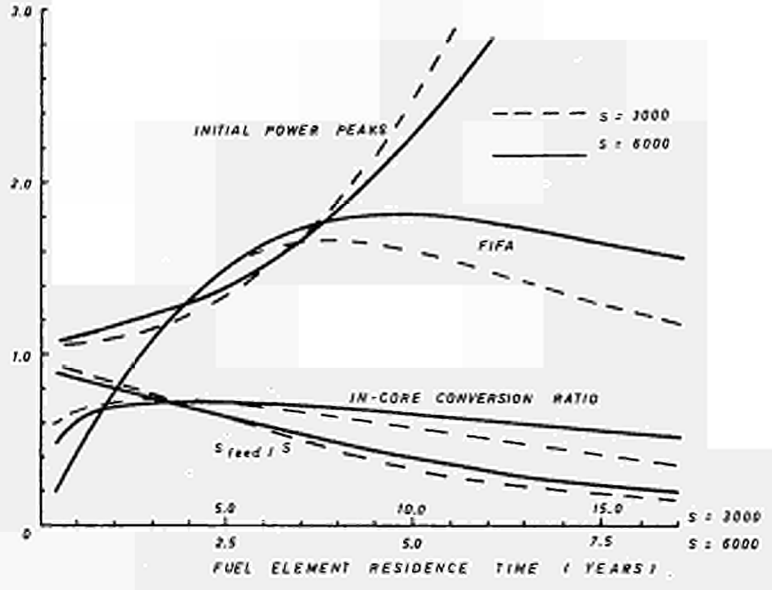
Maximum fifa occurs when :

$$\frac{dN_{U235\ \text{feed}}}{N_{U235\ \text{feed}}} = \frac{dT}{T}$$

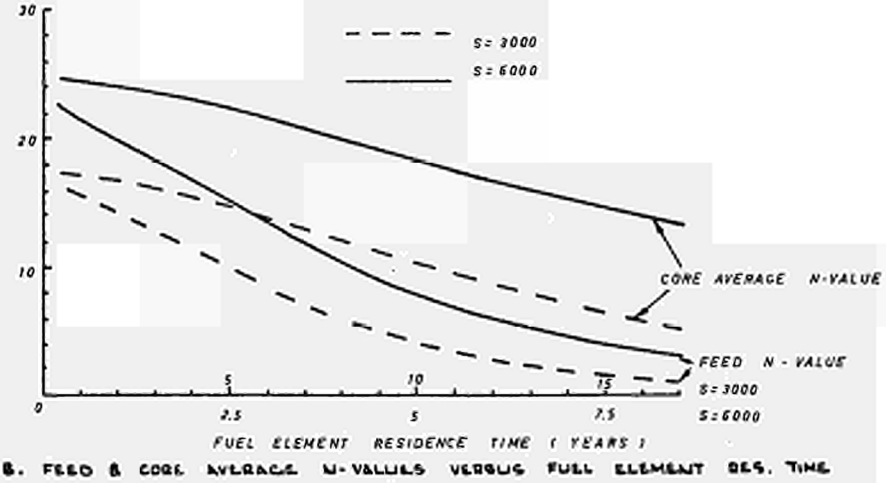
This relation shows also that the power density in the fresh fuel elements increases linearly with running time at maximum fifa.

4.1.2.5. — The Continuous Charge/Discharge Limit.

It is profitable for the understanding of these fuel cycles to study conditions at continuous charge/discharge. We have made calculations on this cycle for power densities between 5 MW/m³ and 15 MW/m³ and S-values between 1,000 and 15,000 over a wide range of fuel element residence times. (In the following S always refers



A. FIFA, CONVERSION RATIO, POWER PEAK & S_{feed}/S VERSUS FULL ELEMENT RES. TIME



B. FEED & CORE AVERAGE N-VALUES VERSUS FULL ELEMENT RES. TIME

FIG. 2. — Continuous charge-discharge analysis of U^{233}/Th once through cycles. 10 MW/m².

to the core average S-value unless otherwise stated). Fuel costs have been evaluated for fuel element fabrication charges between 20£/kg and 200 £/kg heavy metal and for 5 %, 7 % and 10 % interest on capital investment at 100 % load factor.

In order to illustrate the effect of fuel element residence time on fuel cycle performance and fuel costs, we will describe in more detail two systems with the reference design power density 10 MW/m^3 and core average S-values of 6,000 and 3,000.

(*a*) *Fifa, Conversion Ratio, Power Peak,* S_{feed}/S.

Fig. 2 A shows that the overall fuel cycle behaviour of the two cases is very similar, particularly on the left side of the graph, fifa, conversion ratio, fresh fuel power peak and S_{feed}/S depend only on the argument (S $\times$ T). The explicit S-dependence starts to show up at running times corresponding to maximum fifa and above. We can see that for fairly short running times (S $\times$ T $\approx 1.0 \times 10^4$) fifa increases rapidly and the fresh fuel element power peak slowly with increasing fuel element residence time. Later, the trend is reversed and the diagram shows quite clearly how the fresh fuel power density at maximum fifa increases proportionally to running time. Fifa reaches maximum for S $\times$ T values around 2.5×10^4. The conversion ratios go through very flat maxima for (S $\times$ T) values around 1.5×10^4 and decrease very slowly with increasing running time. For practical purposes one can assume the conversion ratio in 10 MW/m^3 systems (assuming 3 % leakage) always to be $\approx$0.7 independent of S and T. The matter will be discussed in more detail below.

It is interesting to note that the ratio between feed and average S-values is an almost linear function in the argument (S $\times$ T) even up to high fifa burn-up values.

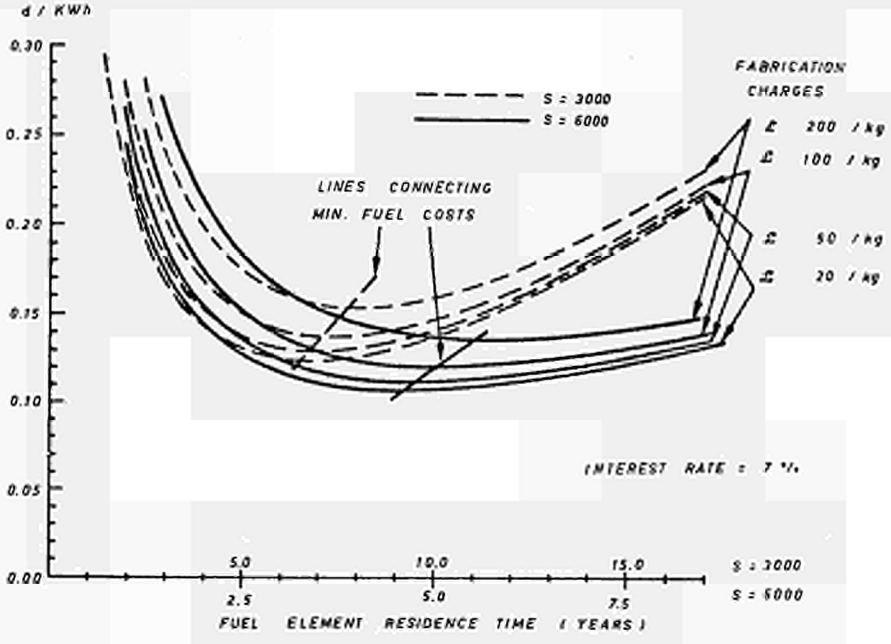
(*b*) *Core Average and Feed N-Values.*

The core average N-value is determined so that the overall conversion ratio shall be 0.7. Since the probability of resonance capture in thorium goes down with increasing S-value this has to be compensated by putting more thorium in the system, thus the core average N-value increases with higher S-values (Fig. 2B). With constant S-value average N-values decrease slightly and feed N-values rapidly with increasing fuel element residence time (because of the corresponding increase in uranium feed).

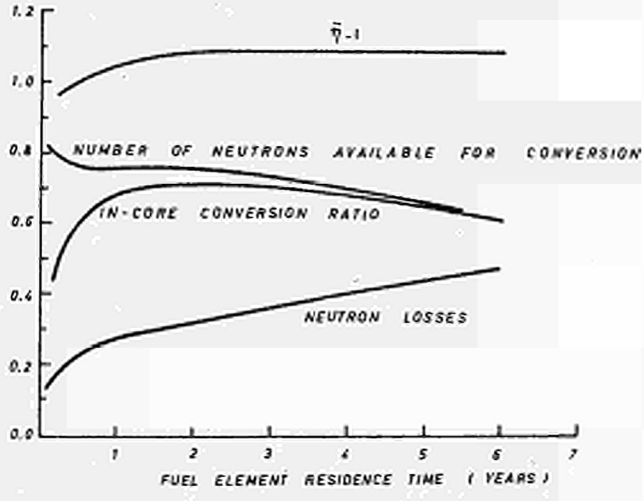
(*c*) *Fuel Costs.*

Fig. 3 A shows fuel costs for the two cases calculated with 7 % interest rate and fabrication charges of 20, 50, 100 and 200 £/kg heavy metal. The lines connecting fuel cost minima for different fabrication charges are drawn. Figs. 2 A and 3 A show that minimum fuel costs are always associated with maximum fifa. In addition it is clear that a reduction in fuel element fabrication charges will allow the fuel element lifetime to be shortened somewhat, which again will lower the power peak and increase the feed N-value.

The overall fuel costs are higher for the undermoderated systems (because of the lower fifa and higher interest charges) and not too much attention should be paid



A. FUEL COSTS VERSUS RES. TIME FOR DIFFERENT FABRICATION CHARGES



B. NEUTRON BALANCE VERSUS RES. TIME S = 6000

FIG. 3. — Continuous charge-discharge analysis of U^{235}/Th once through cycles. 10 MW/m³.

to the fact that fuel cost minima in undermoderated systems imply lower power peaks than in well moderated ones. It should be noted that the cost minima particularly for the S = 6,000 case are very flat indeed. Consequently, if one wants to

reduce the initial power peak by shortening the residence time of the fuel element, this has only a small penalty in fuel costs.

(*d*) *Neutron Balance.*

Returning to the conversion ratio, Fig. 3 B shows the breakdown of this in terms of $\bar{\eta}$-value and non-fertile losses for S = 6,000. We see that at short running times the $\bar{\eta}$-value improves (due to increasing U^{233} content of the system) with the same speed as the fission product poisoning builds up. In this region "the number of neutrons available for conversion" is independent of burn-up. At longer running times the $\bar{\eta}$-value saturates while the fission product poisoning continues to build up, worsening the neutron balance. Further, the difference between the "number of neutrons available for conversion" and the "in-core" conversion ratio becomes larger with shorter fuel element residence times, indeed, going towards zero running times the conversion ratio drops to zero because all protactinium will be discharged. The overall effect is to give a conversion ratio almost independent of burn-up.

This is one of the most interesting features with the once-through cycle, namely that a high fifa burn-up can be obtained without sacrificing the conversion ratio. The penalty in extending the burn-up lies instead in the need to accomodate increasing power peaks in the fresh fuel.

(*e*) *Amount and Composition of Reject Fuel.*

It should be mentioned that if this fuel were to be reprocessed in the future, the uranium reject from the system at maximum fifa would be roughly half of the initial investment. The isotopic composition of this reject uranium would be around 50 % U^{233}, 25 % U^{236}, and the remaining 25 % containing roughly equal amounts of U^{234}, U^{235} and U^{238}.

(*f*) *The Influence of Power Density and S-Value on the Results.*

The fuel cycle at other power densities and S-values shows very similar behaviour. Generally maximum fifa values occur for combinations of P(MW/m^3), S and T (years), defined by :

$$P \times S \times T \approx 2.5 \times 10^5$$

and S_{feed}/S ratios around 0.5.

Fig. 4 shows maximum fifa, maximum conversion ratio and minimum fuel costs (for 50 £/kg fabrication charges at 7 % interest rate) as a function of S-value for different power densities. (The extremes do not occur simultaneously.).

We see that both fifa and conversion ratios improve considerably going towards lower power densities showing the effect of the reduction in protactinium poisoning. Fifa values at constant power densities go through rather flat maxima around S = 6,000, deteriorate at low S-values because of the correspondingly bad η-values for U^{233} and U^{235} and at high S-values due to increased leakage, graphite absorption and Xe^{135} poisoning.

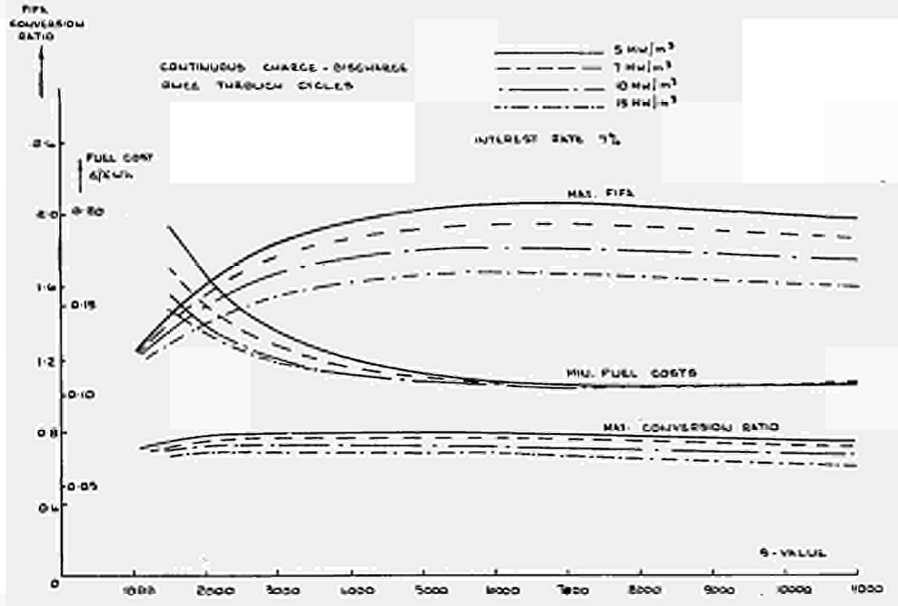


FIG. 4. — Max. FIFA, Max. conversion ratio and min. fuel costs (£50/kg fabrication charges) versus core average S-value for different power densities.

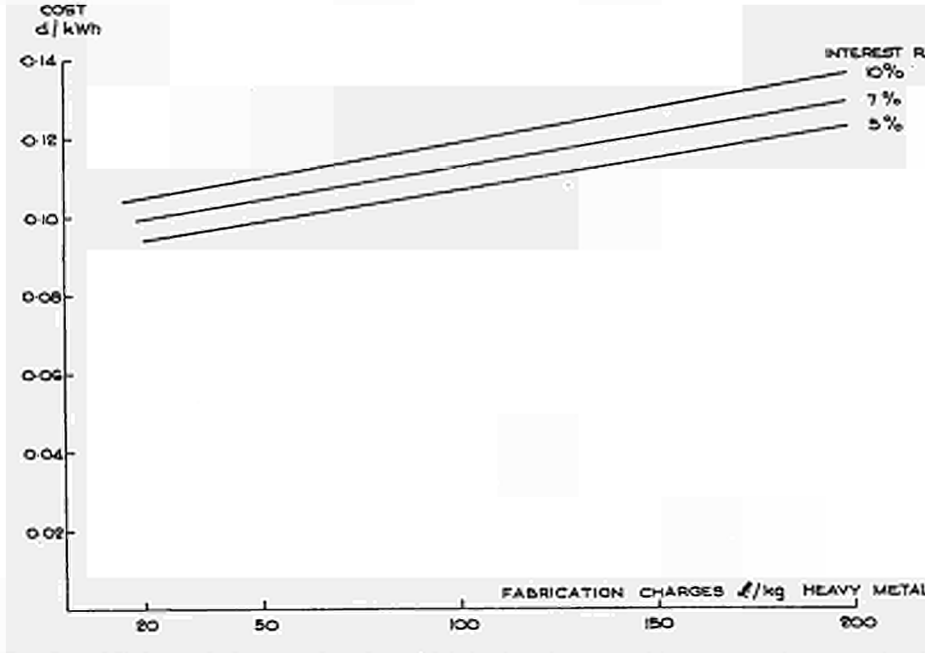


FIG. 5. — Minimum fuel cost as function of fabrication charges and interest rate for power densities between 7 and 15 MW/m³. Continuous charge-discharge. Once through cycles.

However, lowering the power density means increasing the fuel element residence times and increasing the S-value means lowering them. For this reason the better burn-up performance of the low power density systems is offset in fuel costs by the correspondingly higher interest charges; similarly the higher overall fabrication costs of the well moderated systems (because of their higher N-values) are compensated by the lower interest charges. Indeed Fig. 4 shows that above S = 6,000 minimum fuel costs are almost independent of power density and S-value. It is clear

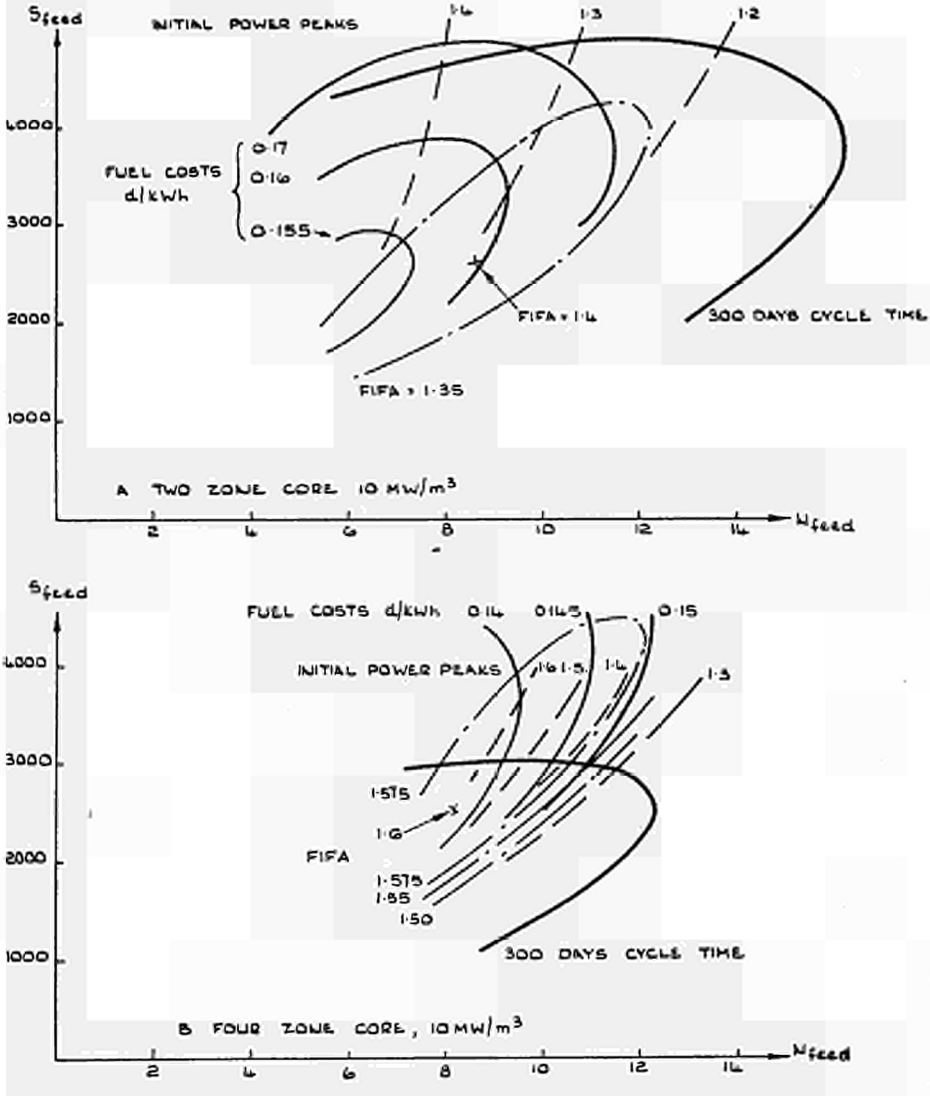


FIG. 6. — FIFA, power peaks and fuel costs (£ 150/kg fabrication charges) in N_{feed}, S_{feed} diagrams annual recharge condition shown. Once through cycles.

that undermoderated systems should not be considered mainly because of their inherent high interest charges but also because of their inferior burn-up behaviour.

The above arguments remain approximately correct for other fabrication charges and slightly different interest rates. Fig. 5 shows minimum fuel costs for the equilibrium continuous charge/discharge once-through cycle as a function of fabrication charges and interest rate for power densities between 7 MW/m³ and 15 MW/m³.

4.1.2.6. — The Effects of Zoning and Cycle Time Restrictions.

It follows from above that if a continuous charge/discharge philosophy were assumed there would be a wide range of possible core arrangements that would satisfy the requirements for minimum fuel costs. However, it is more likely that a reactor system would employ discontinuous loading. The loading periods should preferably coincide with the annual maintenance shut-downs of the plant. In addition the fuel element residence times in the system may be subject to material limitations such as graphite shrinkage, diffusion of active isotopes, etc. This means that we are interested in cores with cycle times of one year and not too many zones.

Detailed studies have been made on systems with 2, 4, 6 and 8 refuelling zones for power densities of 5 MW/m³ and 10 MW/m³. All fuel cost calculations for these systems assume :

Fabrication charges	150 £/kg heavy metal
Load factor	80 %
Interest rate	6 %
Hold-up time of fresh fuel elements	180 days

Figs. 6 and 7 show how the annual recharge requirement determines the core composition for different power densities and zoning arrangements. We have assumed the Reactor to run at full power 300 days every year (corresponding to a load factor ≈ 0.8) and the figures show in (N_{feed}, S_{feed}) diagrams how the line $T_{cycle} = 300$ days intersects the contours for fifa, fuel costs and power peak in the fresh fuel.

Fig.6 A shows that a two-region core with annual recharge on 10 MW/m³ is too far from optimum to be of interest. For the other cases, we find the following results :

Cycle time, years	1	1	1
Power density MW/m³	10	10	5
Number of zones / Fuel element residence time, years	4	6	8
Fifa burn-up	1.60	1.70	1.85
Power peak	1.5	1.8	1.9
Fuel cost, d/kWh	0.143	0.13	0.14
S_{feed}	3,000	2,200	3,500
N_{feed}	10	7	10

The core compositions were determined as to give as low power peaks as possible without too high a penalty in fuel costs.

From these cases we have selected the first one ($S_{feed} = 3{,}000$, $N_{feed} = 10$, $T = 4$ years) for the reference core design because of its relatively low power peak and moderate lifetime in combination with acceptable fuel costs.

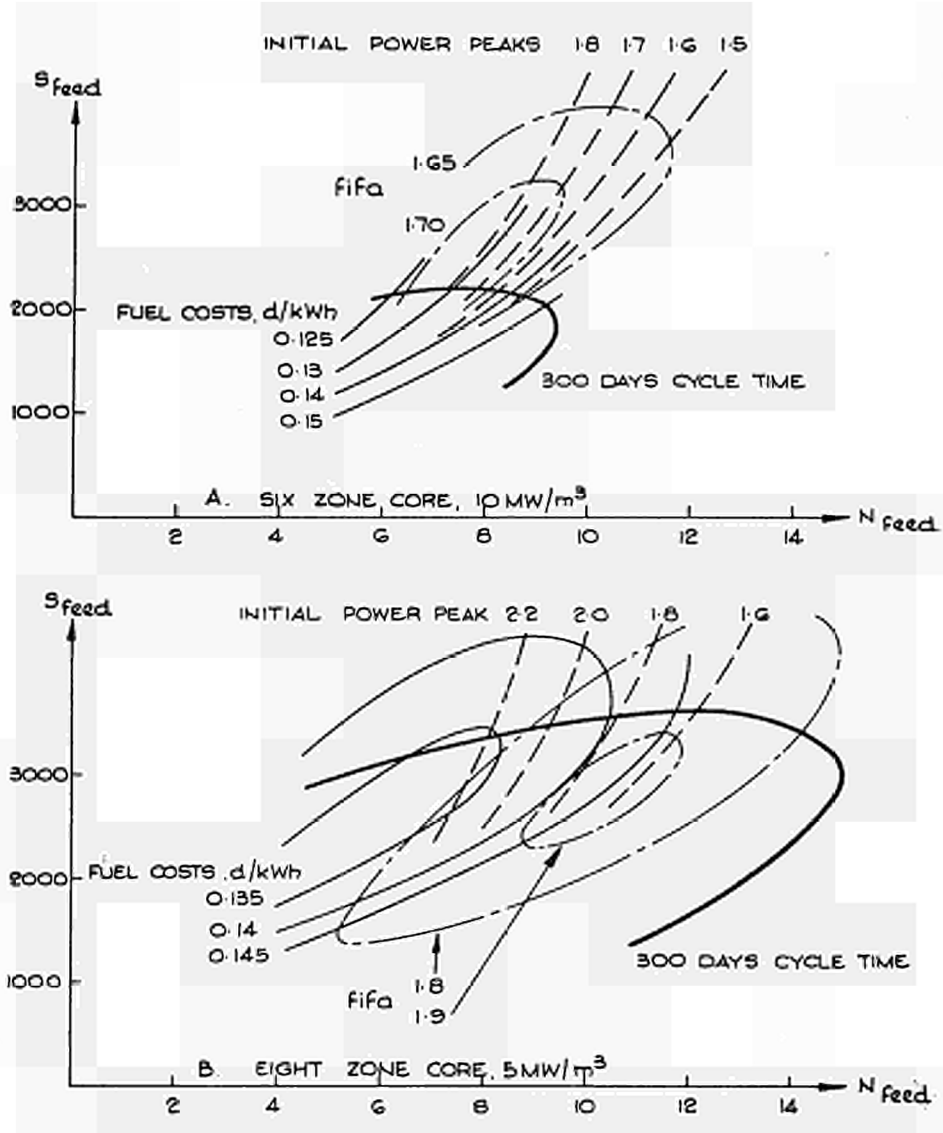


FIG. 7. — FIFA, power peaks and fuel costs (£ 150/kg fabrication charges) in N_{feed}, S_{feed} diagrams. Annual recharge condition shown. Once through cycles.

Fig. 8 shows for 10 MW/m³ and this particular feed composition the fuel cycle behaviour (as discussed in Section 4.1.2.1.) and the fuel costs versus number of zones in the system. The interesting points in this diagram are that :

(i) Above 4 zones the fuel costs saturate rapidly.

(ii) The power peaks in the feed drop with decreasing number of zones.

The latter point follows from the fact that going towards a lower number of zones the feed required to keep the system critical is shared between an increasing number of fuel elements in the core.

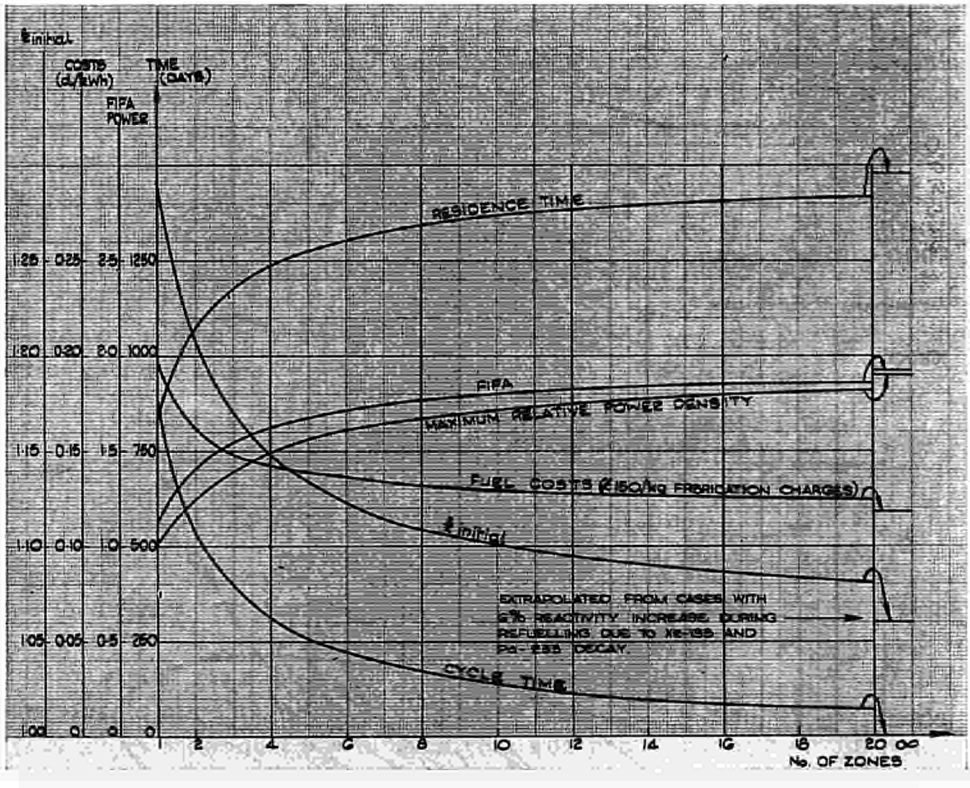


FIG. 8. — Effect of zoning (S_{Feed} = 3,000, N_{Feed} = 10). Once through cycles, 10 MW/m³.

4.2. — SEED BLANKET SYSTEMS.

In a zoned core on the equilibrium cycle U^{235} burns down in a fuel element according to :

$$\frac{N_{U235}}{N_{U235\ initial}} = e^{-\sigma a_{U^{235}} Ft}$$

and U^{233} builds up to equilibrium approximately as :

$$\frac{N_{U233}}{N_{U233\ eq.}} = \left[1 - e^{-\sigma a_{U^{233}} Ft}\right]$$

Since the cross sections for the two isotopes are about the same it follows that U^{233} does not come close to equilibrium until U^{235} has been depleted.

At this moment it would be preferable if the U^{235} and its fission products could be discharged, on the other hand one would like to leave the U^{233} longer in the system to utilise it better and avoid discharging it unnecessarily.

It is clear that on the homogeneous U^{235}/Th cycle these two requirements are contradictory and the residence time of the fuel elements in the system must be a compromise.

We can get away from this by separating the fertile and fissile materials in different fuel elements (blanket and seed elements) with different residence times in the Reactor. It is necessary that these elements are intimately mixed to allow a free exchange of neutrons between them.

4.2.1. — *Physics Aspects.*

In order to investigate the limit to which the physics of the once-through cycle can be improved we looked at a system with only thorium in the blanket and only U^{235} (93 % enriched) in the seed. The analysis was made on an equilibrium cycle assuming continuous charge-discharge fuel handling of *both* seed and blanket elements. This particular cycle is attractive, firstly from the mathematics point of view since it can be treated exactly, secondly it gives a possibility of direct comparison with the homogeneous cycle (which has the identical burn-up performance of a seed-blanket system in which $T_{seed} = T_{blanket}$).

We have looked at a system with an overall power density of 10 MW/m^3 for different S-values over a wide range of residence times for both kinds of elements. The results can be summarised as follows :

In a homogeneous system with given S-value maximum fifa is obtained for a certain fuel element residence time (T_0). Comparison with seed-blanket systems shows that assuming the same S-value we can obtain a 10 % increase in fifa and 7-8 % increase in conversion ratio if :

$$T_{blanket} = 2T_0$$
$$T_{seed} = \frac{2}{3} T_0$$

In the optimum cases the blanket lifetime is thus three times that of the seed. Further about one-third of the total core power will be generated in the seed elements and two-thirds in the blanket. The improved burn-up performance is due to :

(*a*) Immediate discharge of U^{235} fission products once this isotope has been sufficiently depleted.

(*b*) Less U^{233} discharge.

(*c*) Higher overall $\bar{\eta}$-values due to slightly higher $U^{233} : U^{235}$ ratios in the system.

This system has another advantage over the homogeneous cycle; namely that of low U^{236} build-up in the blanket. This is of interest for reprocessing and will be discussed in Section 5.

From a thermal point of view, complete separation of fissile and fertile isotopes has two disadvantages :

(*a*) The initial power density in the blanket elements is zero and builds up slowly with increasing U^{233} content.

(*b*) The ratio of initial to end of life power in the seed is quite high, in the order of 10.

It is beyond the scope of this paper to discuss how the problems in (*b*) can be overcome by separate cooling of the seed and different methods for fuel handling. Constant power in the blanket can be achieved by mixing some U^{235} to the thorium in these elements. This, of course, will worsen the burn-up performance of the system somewhat because of higher fission product build-up in the blanket and slight U^{236} contamination.

4.2.2. — *Economic Aspects.*

The following studies have assumed that the blanket is loaded in the Reactor initially and "primed" with U^{235} so as to give a fairly constant power density during the U^{233} build-up. The seed region is divided into five zones, recharged alternatively. The average power density was assumed to be 5 MW/m^3.

Calculations were made for different initial N-values in the blanket and feed rates of the seed. For the most attractive case ($N_{blanket} \approx 40$) we find after a blanket lifetime of 10 years :

Fifa	=	1.90
Fuel costs	=	0.120 d/kWh
Seed lifetime	=	2.75 years

Leaving the blanket longer in the system will worsen the neutron balance gradually due to the accumulation of fission products. However, the utilisation of the initially invested thorium improves, and we find after 20 years :

Fifa	=	1.92
Fuel costs	=	0.106 d/kWh
Seed lifetime	=	2.20 years

The fuel costs assume 6 % interest rate, 80 % load factor and 150 £/kg heavy metal fabrication charges. Note how the higher fission product poisoning in the blanket after 20 years reflects on the shorter seed lifetime in core and the saturation of fifa. However, the fuel costs drop due to the better thorium utilisation. The lowest fuel costs found for the homogeneous cycle(with the same costing parameters) were 0.135 d/kWh.

The figures quoted here must not be regarded as results from true optimisation studies but yet they show quite nicely the economic advantages of these systems. Generally, the higher the fabrication costs for the blanket, the longer it pays to leave it in the system. In addition , interest charges on the fissile investment in the seed are lower in seed-blanket systems than for homogeneous fuel cycles because of shorter seed lifetime. In conclusion we find seed-blanket concept is attractive both from a physics point of view and for economic reasons.

5. — Fuel cycles assuming reprocessing

We have seen that in a once-through cycle the fuel costs could be optimised solely on a maximum fifa basis without penalty in conversion ratio and this occurred because the $\bar{\eta}$-value of the system improved with burn-up, compensating roughly the increasing neutron losses in fission products. If fuel is reprocessed, however, the feed fuel contains already so much U^{233} that the η-value of the system does no longer increase with-burn-up. Hence the conversion ratio must drop as the fission products accumulate.

Consequently the optimum fuel cycle will be a compromise (based on the costs for reprocessing and refabrication) between the requirements of getting both a high fifa burn-up within each fuel element and also a high return of fissile material from reprocessing it at end of life.

An additional point concerning the use of reprocessed fuel is that handling and refabrication will have to be remote because of the build-up of small quantities of active U^{232} (which has a long chain of active daughters) during irradiation of thorium. This implies higher fabrication charges compared with the once-through cycle.

All fuel cycle calculations reported here refer to a nominal power density of 10 MW/m^3.

5.1. — Homogeneous Fuel.

These calculations assume that U^{235} and thorium are mixed together in the fuel elements of the first cycle of the Reactor to specified values of N_{feed} and S_{feed}. After discharge of a fuel element it is allowed to cool (for Pa^{233} decay) after which uranium is reprocessed (assuming a 2 % loss). Fresh U^{235} make-up and thorium are added during refabrication to give the new fuel element the same feed values of N and S as the initial one. This procedure is repeated after each shut-down.

We have investigated 1- and 4-zone systems on this cycle. We find that even over a total reactor lifetime of 20 years the burn-up performance changes all the time, improving up to about four years after initial start-up because of the fairly rapid saturation of U^{233} in the system, after that deteriorating gradually owing to the accumulation of U^{236} in the fuel. However, these cycles are never very different from the corresponding once-through cycles, therefore, the potential advantage of reprocessing the U^{233} for its high η-value is offset by the poisoning of U^{236}.

Cost evaluations (assuming 6 % interest and 80 % load factor) have been made over a wide range of fabrication and reprocessing charges. The sum of these two items is the important quantity that determines the nuclear design for minimum fuel costs. Generally, the lowest fuel costs occur for core average S-values around 6,000.

It is beyond the scope of this paper to discuss in detail the time behaviour of the fuel cycle over the Reactor life. Fig. 9 illustrates how the sum of fabrication and reprocessing charges feeds back on conversion ratio, make-up and fifa (1- and 4-zone cores) and power peak (4-zone core) for minimum fuel cost systems. The values shown are those obtained after a few years' running time when the fuel cycle performs best and before too much U^{236} has accumulated in the system. (The make-up is defined as the fraction of purchased U^{235} in the fissile feed of a fuel element.) We see how the conversion ratio increases and fifa, make-up and power peaks drop with lower fabrication and reprocessing charges.

Fig. 10 shows minimum fuel costs for 1- and 4-zone cores versus fabrication plus reprocessing charges. The point of lowest fuel costs in a 4-zone core on the

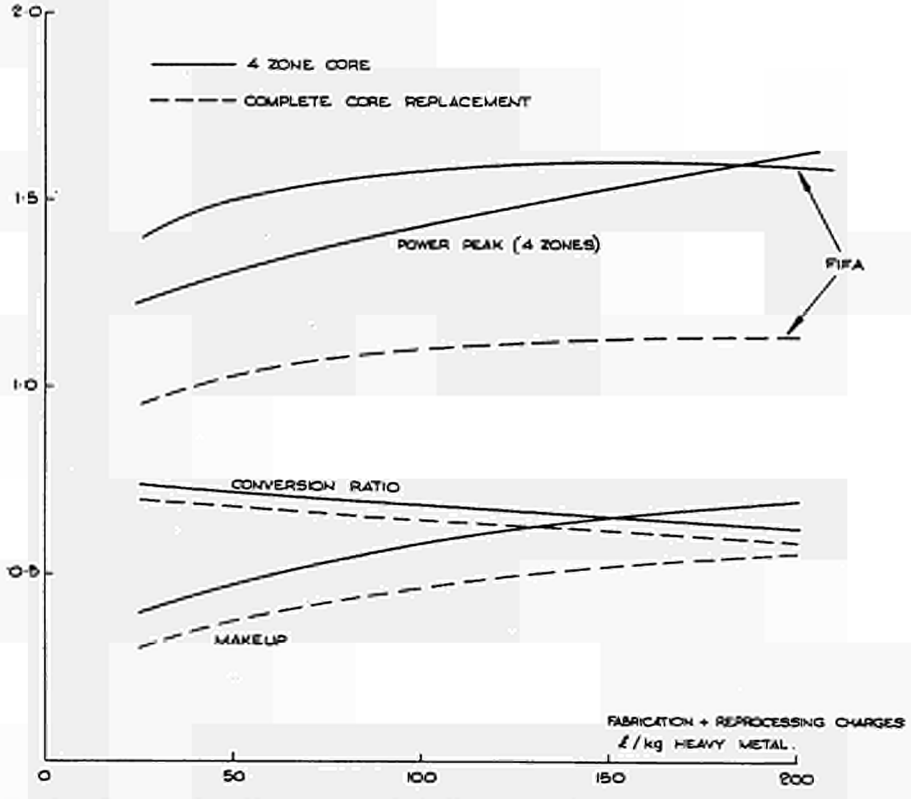


FIG. 9. — Reprocessing of homogeneous fuel. FIFA conversion ratio, makeup and power peak versus fabrication + reprocessing charges at minimum fuel costs. 10 MW/m³.

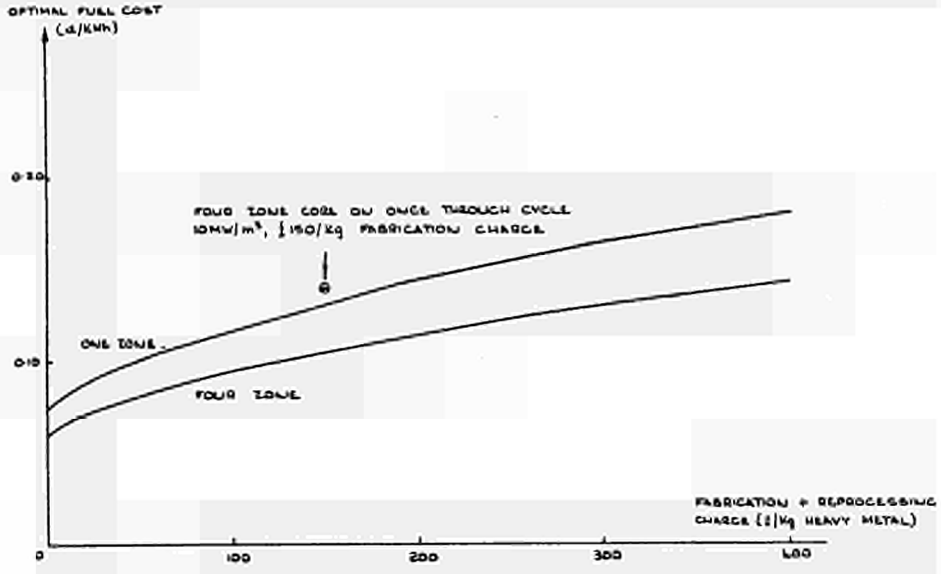


FIG. 10. — Reprocessing of homogeneous fuel. Optimised fuel costs versus fabrication + reprocessing charges. 10 MW/m³.

once-through cycle (with 150 £/kg fabrication charges) is shown. The same fuel costs are obtained for the 4-zone core on a reprocessing cycle for fabrication plus reprocessing charges of 370 £/kg. Thus break even between the once-through cycle and the reprocessed fuel cycle occurs when the cost of reprocessing and the additional cost of remote refabrication is 220 £/kg heavy metal.

5.2. — SEGREGATED FUEL.

We have seen that fuel cycles, based on reprocessing of homogeneous fuel do not lead to markedly better neutron economy or burn-up performance than the once-through cycles because of U^{236} build-up. In order to take full advantage of the superior characteristics of U^{233} as fuel it is necessary to arrange the fuel cycle in such a way as to prevent U^{236} from accumulating in the system.

Since this isotope is built up from neutron captures in U^{235} its poisoning influence on the fuel cycle can be avoided by keeping the make-up fuel separate from the thorium and discharging it after depletion. In a seed-blanket system this would be automatically achieved.

Reprocessing of the thorium (the blanket) after discharge will yield uranium rich in U^{233} and with a very low U^{236} content.

5.2.1. — *Different Ways of Arranging the Fuel Segregation — The Particular Cycle Investigated.*

One would be perfectly free to reinvest the recovered uranium either into a new blanket or into seed fuel. Putting it in the blanket would hardly be disadvanta-

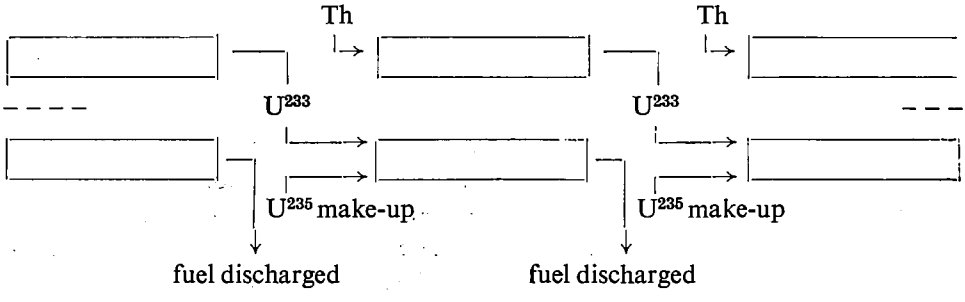
geous from a neutron balance point of view since neutron captures in U^{236} produced from thorium will be less than a factor :

$$\frac{\alpha_{U\,233}}{1+\alpha_{U\,233}} \times \frac{\alpha_{U\,235}}{1+\alpha_{U\,235}} \approx 0.02$$

of the conversion ratio. This formula represents the probability of forming U^{234} out of U^{233} times the probability of forming U^{236} out of U^{235} (the probability of forming U^{235} out of U^{234} is 1).

However, mixing active U^{233} to non-active thorium will necessitate remote refabrication of the whole blanket. Mixing it instead to the make-up U^{235} will imply remote fabrication only of the seed.

We have investigated the latter cycle with the restriction that fissile and fertile materials are kept for the same times in the system but segregated physically. The analysis was made assuming continuous charge/discharge fuel handling and the following fuel management scheme :



We have studied cases for a power density of 10 MW/m³ and S-values between 3,000 and 8,000 over a wide range of fuel element residence times.

5.2.2. — *Results.*

Results for the equilibrium condition are given in Figs. 11 and 12 for S = 3,000 and S = 6,000.

(*a*) *Fifa, Conversion Ratio, Power Peak,* S_{feed}/S, *Make-up.*

Fig. 11 A shows fifa, in-core conversion ratio, power peak in the fresh fuel, S_{feed}/S and fraction of make-up in the feed versus fuel element residence time. (The power peaks assume that fissile and fertile fuel is kept in the same fuel element and refer to the overall heat production in the element initially).

Like the corresponding cases in the once-through cycle, Fig. 6 A, these quantities apparently depend also mainly on the product (S × T). Compared to the once-through cycles the conversion ratios are considerably higher and have definite maxima coinciding with minimum make-up. Fifa values are higher and power peaks lower than in the once-through cases; these are consequences of the better fuel utilisation.

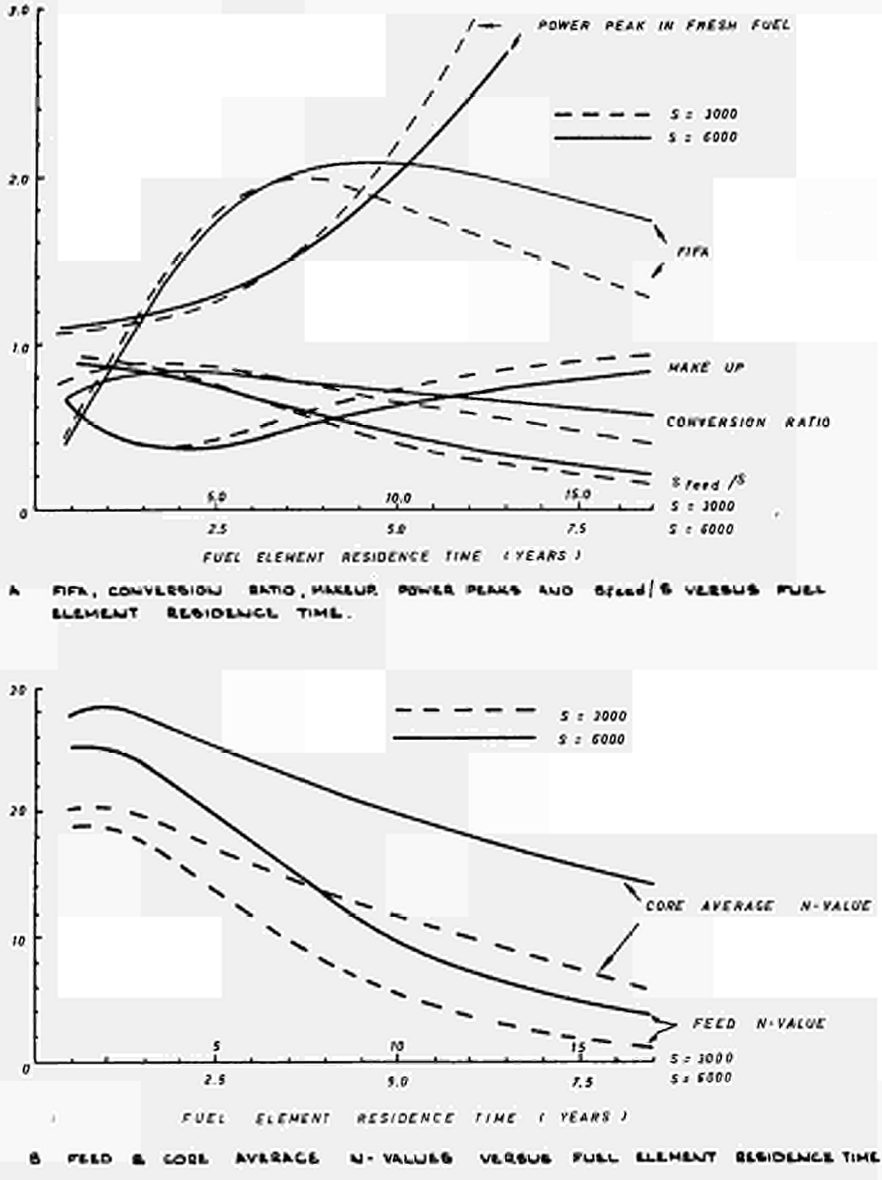
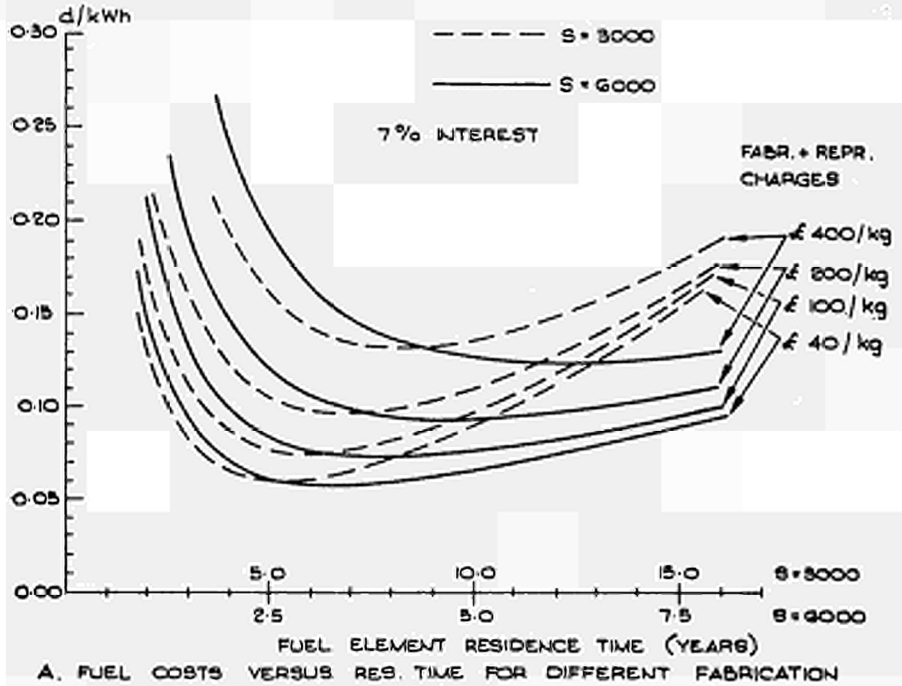
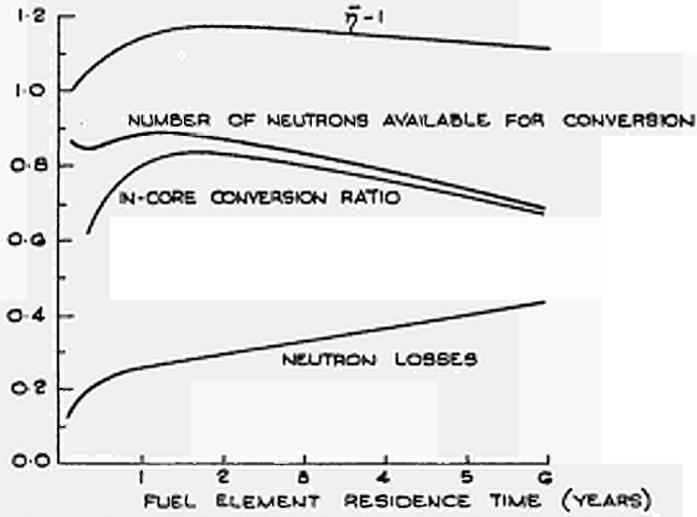


FIG. 11. — Continuous charge-discharge analysis of U^{233}/Th fuel cycles assuming reprocessing. Segregated fuel, 10 MW/m³.

The fact that the make-up goes through a definite minimum depends on the particular fuel management scheme investigated. Going towards very short fuel element residence times the fissile fuel will not stay in the core long enough for sufficient depletion and will instead be lost at discharge. More make-up material must then be added to account for this loss.



A. FUEL COSTS VERSUS RES. TIME FOR DIFFERENT FABRICATION + REPROCESSING CHARGES.



B. NEUTRON BALANCE VERSUS RES. TIME. S = 6000

FIG. 12. — Continuous charge-discharge analysis of U^{233}/Th fuel cycles assuming reprocessing Segregated fuel, 10 MW/m³.

(*b*) *Feed and Average N-Values.*

Fig. 11 B shows core average and feed N-values. The influence of the conversion ratio on these quantities can be clearly seen. Note how sharply the feed N-value falls with increasing fuel element residence times.

(*c*) *Fuel Costs.*

Fig. 12 A shows that minimum fuel costs always occur in the region of maximum fifa. When fabrication and reprocessing charges are high, cost minima occur for low N-values implying long fuel element residence times and correspondingly high initial power peaks. If these charges are low there will instead be an incentive to have as low make-up as possible implying relatively short fuel element residence times and moderate power peaks. Only in latter cases will one obtain high conversion.

(*d*) *Neutron Balance.*

Fig. 12 B shows the neutron balance for S = 6,000. We see how the $\bar{\eta}$-value is essentially constant, decreasing slightly towards low and high fuel element residence times because of the higher U^{235} content in these systems. The neutron losses increase steadily with residence time due to fission product accumulation and the conversion ratio drops accordingly. In the once-through cases (Figs. 7 B and 7 C) we concluded that the in-core conversion ratio was the important one since Pa^{233} decays in a fuel element after having left the system are of no benefit for the fuel cycle. On a reprocessing cycle however, all uranium will be recovered regardless of whether it was produced by Pa^{233} decays in the Reactor or out of it. This gives a better neutron utilisation and the true conversion ratio is here equivalent to "the number of neutrons available for conversion."

It is interesting to see how the effects of $\bar{\eta}$, neutron losses and neutron utilisation all contribute to give higher conversion ratios for reprocessed fuel on a segregated cycle than for once-through systems. Clearly, the use of reprocessed fuel will result in higher $U^{233} : U^{235}$ ratios giving higher overall $\bar{\eta}$-values and lower U^{236} losses. In addition the yields for the most important fission products are generally lower for U^{233} than for U^{235}. Comparison between Figs. 7 B and 12 B show the higher $\bar{\eta}$-values and lower losses in the system using reprocessed fuel.

5.2.3. — *Comparison of Fuel Costs with Once-Through Cycle.*

Turning to the fuel costs Fig. 13 shows these as a function of fabrication and reprocessing charges at 7 % interest rate and load factor 1. In the same diagram the corresponding costs for the once-through cases are given versus fabrication charges. The curves are almost parallel and show that on continuous charge-discharge break even between the homogeneous once-through cycle and the particular segregated fuel cycle considered occurs for reprocessing charges of $\approx$ 220 £/kg heavy metal (including the additional charges for remote refabrication of the fissile

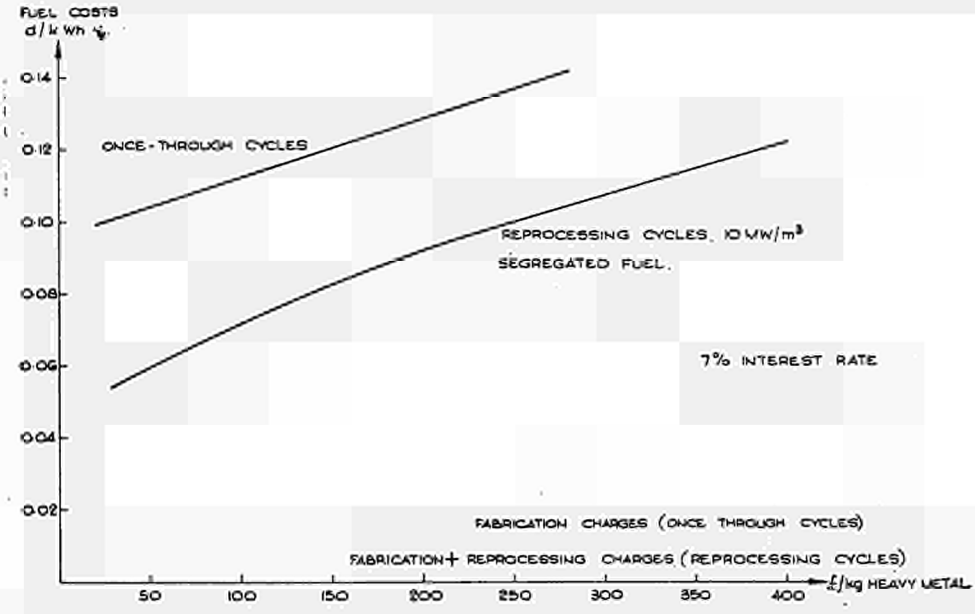


FIG. 13. — Minimum fuel costs. Comparison between once-through and reprocessing cycles continuous charge-discharge fuel handling.

fuel) independent of fabrication costs. Generally, the feedback of fabrication and reprocessing charges on fuel costs and core design is much stronger for cycles assuming reprocessing than for the once-through systems because the contribution of these costs to the overall fuel costs is larger.

5.2.4. — *Additional Remarks.*

We could see in Fig. 11 B that the overall N-values increase with the S-value of the system. Clearly, the turnover of heavy metal per kg U^{233} going through reprocessing and refabrication is reduced by going to undermoderated systems because of their inherent lower N-values, and this effect is much more important here than on the once-through cycle (where the higher interest charges compensated this effect on the fuel costs for $S \geqq 5{,}000$). Indeed we find that the fuel costs are almost the same for S-values down to 3,000 (Fig. 12 A) and consequently one can go further towards undermoderated systems if fuel is reprocessed than on the once-through cycle.

These calculations were mainly intended to illustrate the influence of segregation and reprocessing on the fuel cycle and the fuel costs. It is very likely that even better fuel cycle performance can be obtained with different segregation schemes, particularly if the fissile and fertile materials are allowed different residence times in the system. Such fuel cycles have yet to be investigated.

6. — The sensitivity of the Results to change in Neutron Leakage, Temperature and Graphite Absorption

We have investigated the stability of our results against changes in the parameters assumed in Section 3.1 on the once-through cycle assuming continuous charge/discharge fuel management.

(i) Variations in moderator and fuel temperatures have negligible influence on fifa and fuel costs (at constant thermal efficiency). This is understandable since the thermal α-values for both U^{235} and U^{233} are almost independent of neutron energy. However, lower temperatures tend to give slightly higher N-values.

(ii) At S-values around 6,000 (optimum fuel cycle performance) the overall graphite absorption is relatively small. The influence on the fuel cycle performance of graphite impurities or thermal flux depression in the fuel was found to be insignificant.

(iii) Small variations in the graphite content of the core will not change the burn-up performance or fuel costs for given fuel ratings. The S-values will change proportionally to the number of carbon atoms per unit core volume.

Similarly the N-values will increase with S-value to compensate for the decreased resonance absorption.

(iv) Changes in leakage rate have a very significant influence on the burn-up performance. Fig. 14 shows how maximum values of fifa and conversion ratio change with leakage rate at an S-value of 6,000 and power densities of 5 MW/m³

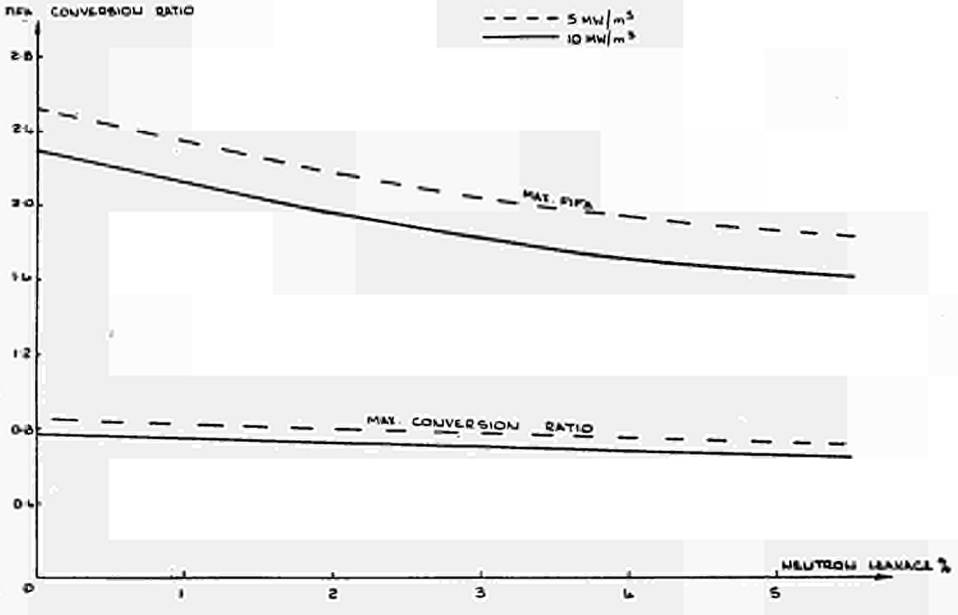


Fig. 14. — Leakage dependence of FIFA and conversion ratio. Continuous charge-discharge once through cycles. S = 6,000.

and 10 MW/m³. The improvement in burn-up performance that can be obtained by cutting the leakage losses is rather striking. Doing this in practice would probably be very difficult since it would imply going to very large core dimensions.

(v) The assumption of homogeneous fuel will lead to slight underestimates of the N-values, particularly for the undermoderated systems.

7. — Conclusions

We have seen that on a once-through cycle with homogeneous fuel :

(i) The minimum fuel costs are practically independent of power density.

(ii) The main cost item is the U^{235} charge but fuel fabrication costs play an important part. However, even with very low fabrication costs it would be difficult on this cycle to come below 0.12 d/kWh, particularly if one is restricted to annual recharge.

(iii) Recharging in parts of the core is imperative for obtaining low fuel costs but implies high power peaks in the fresh fuel elements.

(iv) Undermoderated systems (core average S-values less than 5,000) are not interesting.

Separation of fissile and fertile material into seed and blanket will give :

(i) Better utilisation of thorium.

(ii) Less discharge of fissile material from the system.

(iii) Better neutron balance.

(iv) Lower fuel costs (down to 0.10 d/kWh).

Fuel reprocessing is of interest if it could be made cheaper than 220 £/kg heavy metal. We find for homogeneous fuel on reprocessing cycles :

(i) The fuel cycle performance is similar to and not markedly better than the once-through cycles.

(ii) The fuel costs will still be lower but the improvement is marginal (down to 0.10 d/kWh) with annual recharge.

If a fuel segregation scheme is adopted this will lead to :

(i) Higher conversion and better fuel utilisation.

(ii) Undermoderated systems are of interest on this cycle down to core average S-values of 3,000.

(iii) Fuel costs become very dependent on fabrication and reprocessing costs. Our very preliminary investigations show that if the sum of fabrication and reprocessing charges is 100 £/kg, fuel costs of 0.07 d/kWh can be obtained.

It must be realised that even though our burn-up calculations are quite accurate the absolute value of the fuel costs is uncertain for lack of knowledge of the U^{235} price. Further, we are probably conservative in assuming that fabrication and reprocessing costs depend only on the quantity of heavy metal to be handled without regard to composition. If the factor determining these costs were instead the fissile content of the throughput (batch sizes in the handling being determined by criticality considerations, for instance) the penalty in going to higher N-values would

be smaller. This would change the nuclear design towards better moderated systems and shorter fuel element lifetimes.

ACKNOWLEDGMENTS

The authors wish to acknowledge Dr. K. O. Hintermann and Dr. E. Schröder for their continuous interest and support and thank Miss S. Smith and Mr. M. White for their assistance in carrying out the numerical calculations.

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APPENDIX

LIST OF DEFINITIONS

S : Atomic density ratio between graphite and fissile isotopes (U^{233}, U^{235}, Pu^{239}, Pu^{241}) in the equilibrium system.

S_{feed} : The same ratio relating to the initial composition of a fuel element.

N : Atomic density ratio between thorium and fissile isotopes in the equilibrium system.

N_{feed}; The same ratio relating to the initial composition of a fuel element.

Fifa : The ratio of total number of fissions in a fuel element to the number of fissile atoms originally invested.

Conversion Ratio :

(*a*) Number of neutrons available for conversion = total number of neutrons produced minus fissile and non-fertile losses.

(*b*) In-core conversion ratio = the ratio between the rates of production and destruction of fissile material in the core.

The difference between these quantities account for Pa^{233} discharge from the system.

Make-up : The fraction of fissile material in a fresh fuel element that has been purchased externally.

η : Number of fission neutrons per fissile absorption in a particular isotope.

$\bar{\eta}$: Average number of fission neutrons per fissile absorption in a system containing several fissile isotopes.

Compound Interest : Annual capitalisation.

(1) Dragon Project Internal Report : available only to authorized persons and firms in the ount ries participating in the Dragon Agreement.

FUEL-CYCLE COST COMPARISONS FOR HIGH-TEMPERATURE GAS-COOLED REACTOR FUELS (1)

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ABSTRACT

Calculations were made of the burnup, conversion ratio, and fuel-cycle cost over a wide range of fuel compositions and fuel management modes for semi-homogeneous uranium and uranium-thorium fuels. A limited series of heterogeneous lattices was also investigated in connection with low-enrichment uranium fuel. Lowest fuel-cycle cost [0.88 mills/kWhr(e)] for an economy containing large-scale low-cost fabrication and reprocessing facilities was found to be the uranium-thorium recycled fuel. For near-term economics with high fuel fabrication costs, lower fuel-cycle costs may be obtained from a semi-homogeneous partially enriched uranium fuel operated on a throw-away cycle. We have not been able to find any advantage to a heterogeneous low-enrichment uranium fuel cycle in a high-temperature gas-cooled reactor.

1. — SEMI-HOMOGENEOUS GRAPHITE-MODERATED FUELS

We have made calculations of the reactivity lifetimes and the associated conversion ratios and fuel-cycle costs for a wide variety of fuel compositions that might be used in a high-temperature gas-cooled reactor. The results for fully enriched uranium with thorium and for partially enriched uranium are described in this paper. Parallel calculations have also been made for plutonium-thorium fuels, and these results are discussed elsewhere [1].

For each type of fuel we varied the moderator-to-fuel ratio and fissile-to-fertile ratio and computed the reactivity lifetime. All of the calculations were made for the slightly idealized case of a graded-exposure equilibrium fuel cycle in which the fuel was assumed to be fed continuously into the reactor and discharged at some later time, also continuously. The calculated neutron spectrum for all fuel elements was characteristic of the average composition and did not change with time. Two different types of fuel management were considered. In non-recycle fuel management it was assumed that the fuel discharged from the reactor was to be reprocessed and

(1) ORNL-report TM-1112, April 1965.

(2) Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

TABLE I. — Fixed Parameters for Fuel-Cycle Calculation

Power density, w/cm³	5.0
Thermal efficiency, %	40
Reactor plant factor	0.8
Average moderator temperature, °K	900
Buckling, cm^{-2}	2.0×10^{-5}
Isotopic composition of fully enriched uranium, wt % :	
U^{234}	1.04
U^{235}	93.15
U^{236}	0.22
U^{238}	5.59
Graphite matrix density, g/cm³	1.65
Coolant fraction	0.39
Fixed charges on fuel inventory, %/year	10
Fixed charges on working capital, %/year	10
Fabrication holdup time, days	150
Processing holdup time, days	150
Processing losses, % :	
Uranium and plutonium	1.0
Protactinium	3.1
Fabrication scrap losses, %	0.2
Fuel shipping charges, $/kg of heavy metal :	
To processing plant	6.40
From processing plant	5.05
Cost of U^{235}, $/g [a]	12.05
Cost of U^{233}, $/g [a]	12.05
Cost of plutonium, $/g fissile	10.0

[a] This price is for fully enriched material. The USAEC cost schedule was used for lower enrichments.

sold or discarded without reprocessing. In the other type, recycle fuel management, it was assumed that the uranium and plutonium discharged from the reactor were to be reused after reprocessing along with sufficient makeup fuel to maintain the required fissile loading. The isotopic concentrations of the recycled material were calculated to meet the condition that the quantity of each isotope discharged, less the processing losses, should be equal to the quantity fed from the recycle stream. Additional makeup fuel of a specified composition was fed as required.

The lifetimes were computed with a space-independent code in which the leakage was allowed for by the inclusion of a buckling term giving 2.0 to 2.5 % neutron leakage. The moderator was carbon (graphite) in all cases, and it was assumed that the moderator and fuel were essentially homogeneous with respect to neutron behavior. The calculations were done with 11 fast and 20 thermal energy groups. Fission-product concentrations were calculated explicitly for the 35 most important nuclides, and a pseudoelement was used for the remainder of the fission products.

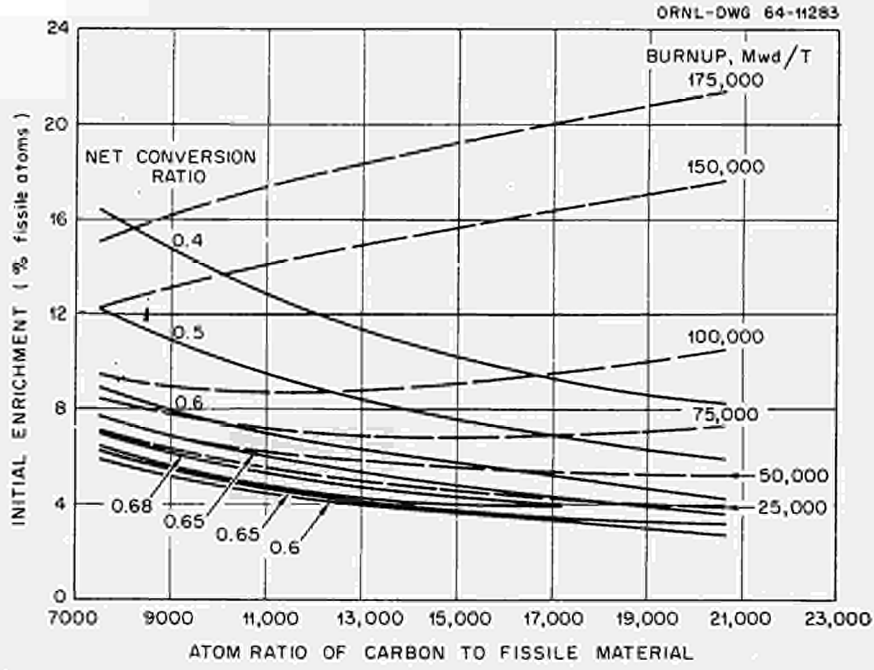

FIG. 1. — Burnup and Conversion Ratios for Recycled Partially-Enriched Uranium Fuel.

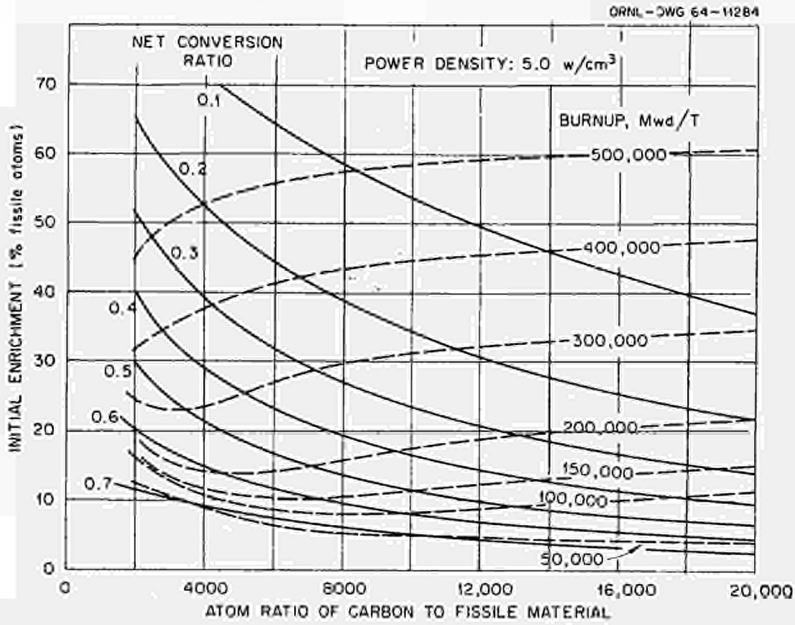

FIG. 2. — Burnup and Conversion Ratios for Non-Recycled Partially-Enriched Uranium Fuel.

Earlier studies [2] have indicated that minimum fuel-cycle costs for the U^{235}-thorium fuel are to be found with a power density of 5-10 w/cm^3. The minimum occurs as a result of the contrasting effects of greater parasitic captures in Xe^{135} and Pa ^{233} at high power density and larger fuel inventory costs at low power density. In this study we did not vary the power density : a value of 5 w/cm^3 was used for all cases. Some of the other assumptions made in the study are given in Table I.

The conversion ratios and reactivity lifetimes that were obtained are shown in Figs. 1 through 4 as functions of the fuel feed composition. The highest conversion ratios were obtained for the thorium-fully enriched uranium fuel with recycle (Fig. 3). In these cases the conversion ratio can be above 0.9 for burnups of 25,000-30,000 Mwd/T and can approach 1.0 for very short burnup. It should be emphasized that these calculations did not allow for such devices as selected partial recycle, fertile blankets or removal of fission products during irradiation. Such devices could increase the conversion ratio for economically attractive cycles to near unity or perhaps even higher. In the recycled thorium-uranium fuels with high conversion ratio nearly all of the fissions occur in the bred U^{233}, and it is the high η value of the U^{233} which is primarily responsible for the superior nuclear performance of these fuels. A concurrent disadvantage of the recycled fuel is the buildup of U^{236} in the recycled stream which results in neutron absorption in both U^{236} and Np^{237}. As the conversion ratio is decreased, the proportion of U^{235} makeup in the recycled fuel becomes greater, and the buildup of U^{236} increases. Consequently, at conversion ratios less than about 0.75 the burnup at a given conversion ratio is greater with non-recycled uranium-thorium fuel (Fig. 4) than with recycled uranium-thorium fuel (Fig. 3). The conversion ratios obtained with partially enriched uranium fuels were never greater than about 0.7 at the high burnups required for an economical cycle with this type of fuel. At a given burnup the conversion ratios tended to be lower for the recycled than for the non-recycled partially enriched uranium. The combination of high burnup and low conversion ratio in the higher-enrichment fuels gave a very great depletion of the fissile material in a fuel element.

The various assumptions regarding economic parameters are given in Table I and in Fig. 5. It should be noted in Fig. 5 that the costs per unit weight of heavy metal for fabrication and for reprocessing were allowed to vary with throughput. These costs were recently calculated as a part of a comprehensive study of the probable reactor industry in the period following 1970 [3]. An industry size of 15,000 Mw(e) was assumed. We used remote fabrication for all recycled fuel, hooded fabrication for the thorium-fully enriched fuel, and direct fabrication for the partially enriched uranium fuel. The cost of U^{235} of various enrichments follows the current USAEC schedule and appears to be a reasonable projection for a period some time in the future. In mixtures containing U^{233}, the U^{233} was treated like U^{235} which results in a slight under evaluation for most cases. A 10 % interest rate was used both for fuel inventory and for working capital for fuel fabrication; this rate is considered representative for private financing.

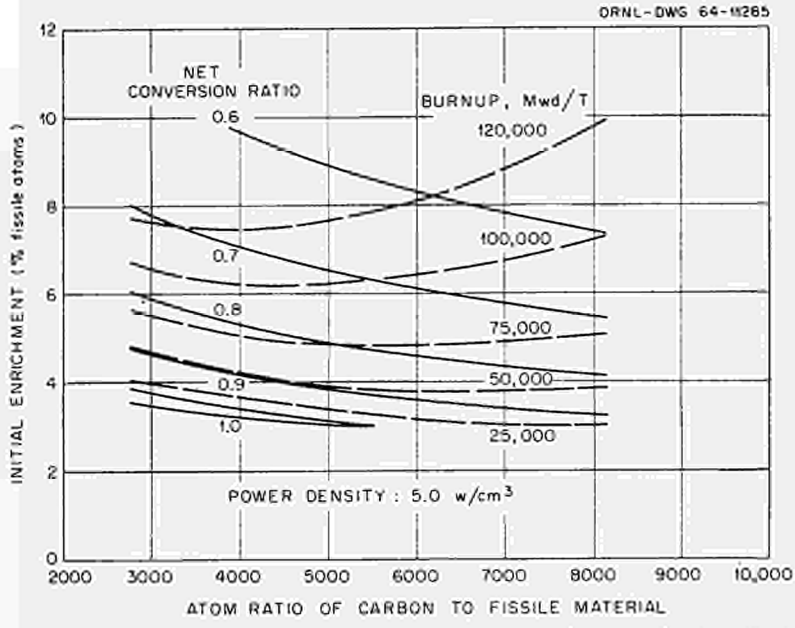

FIG. 3. — Burnup and Conversion Ratios for Recycled Uranium-Thorium Fuel.

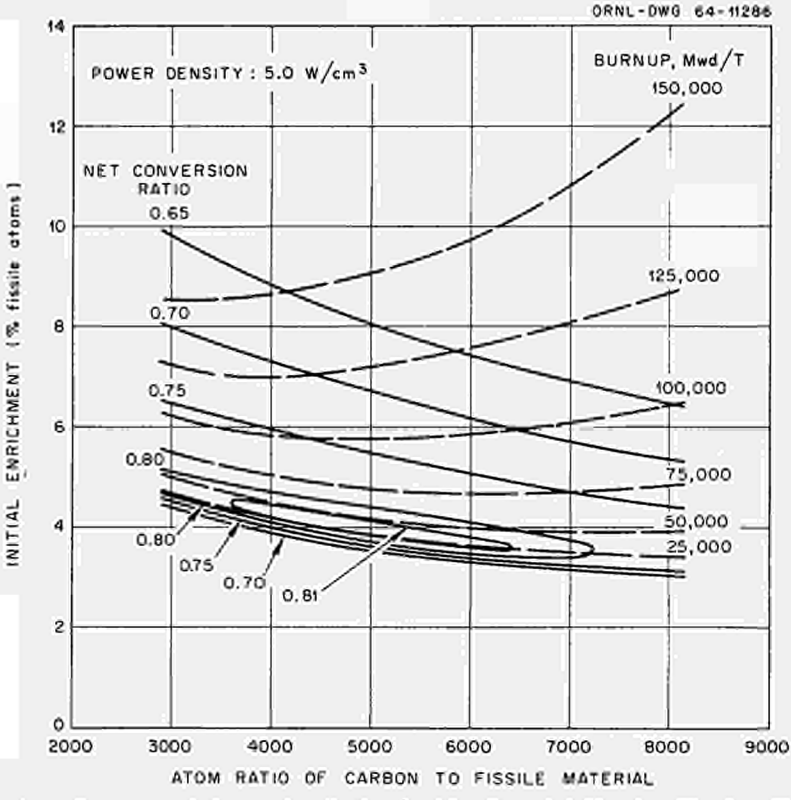

FIG. 4. — Burnup and Conversion Ratios for Non-Recycled Uranium-Thorium Fuel.

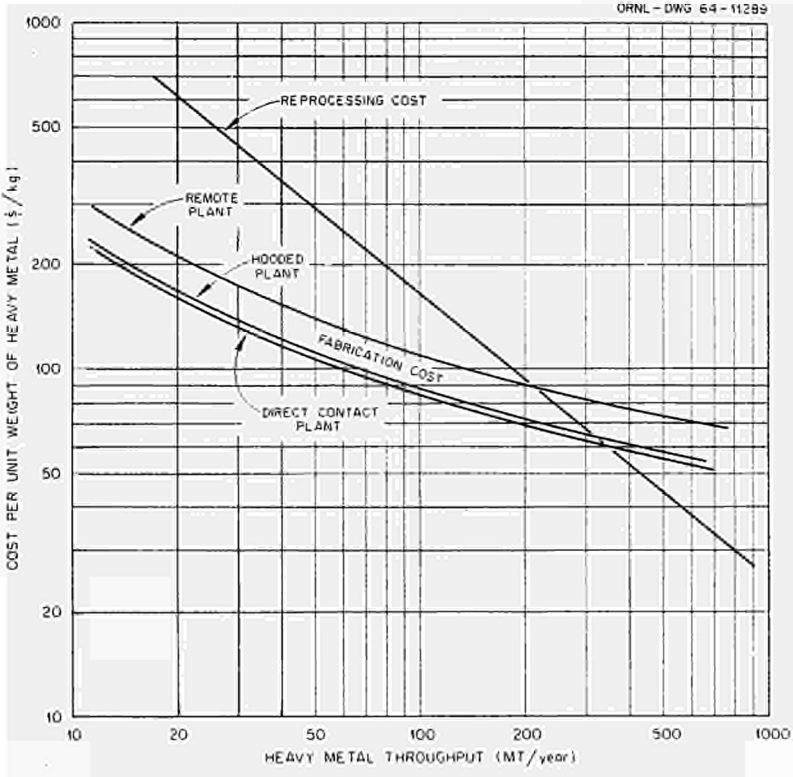

FIG. 5. — Unit Cost Estimates for Fabrication and Reprocessing.

TABLE II. — Minimum Fuel-Cycle Cost for Uranium-Thorium and Partially Enriched Uranium Homogeneous Fuels

	Minimum Fuel-Cycle Cost, [mills/kwhr(e)]			
	Partially Enriched Uranium Fuel		Uranium-Thorium Fuel	
	Not Recycled	Recycled	Not Recycled	Recycled
Reprocessing	0.155	0.172	0.159	0.177
Fabrication	0.085	0.163	0.098	0.183
Fabrication interest	0.008	0.011	0.014	0.022
Shipping	0.006	0.019	0.007	0.023
Uranium feed	0.740	0.680	0.767	0.219
Uranium credit	(0.021)		(0.334)	
Plutonium credit	(0.100)		(0.000)	
Core inventory	0.057	0.088	0.128	0.189
Fabrication inventory	0.030	0.048	0.032	0.038
Processing inventory	0.005	0.022	0.014	0.029
TOTAL	0.965	1.203	0.885	0.880
Total with spent fuel discarded [a]	0.926		1.013	

[a] Fuel compositions were re-optimized in computing the costs on this line.

The fuel-cycle costs are tabulated in Table II for the composition that gave the lowest total cost with each type of fuel. Other data for these same cases are given in Table III and Table IV.

The total fuel-cycle costs including fuel reprocessing are significantly lower for the thorium-fully-enriched uranium fuels than for the partially enriched uranium fuels. The thorium-based fuel gives a cost of 0.88 mills/kwhr(e) for the recycled mixture and essentially the same total when the spent fuel is reprocessed and sold. The partially enriched uranium fuel gives a cost of 1.20 mill/kwhr (e) when the fuel is recycled and 0.96 when the fuel is reprocessed and sold. The better cost of the thorium-based fuel comes principally from the higher conversion ratios that occur when most of the fissions are in U^{233}, leading to a much lower net cost of fissionable material. The recycled uranium-thorium fuel has the lowest fuel makeup cost and the lowest total fuel cycle cost in spite of high fabrication cost owing to the necessity of remote fabrication.

TABLE III. — Homogeneous Fuel Compositions Yielding Minimum Cost

	Partially Enriched Uranium Fuel		Uranium-Thorium Fuel	
	Not Recycled	Recycled	Not Recycled	Recycled
Fresh fuel composition :				
Moderator-to-fissile atom ratio .	10,000	10,000	7,000	7,000
Enrichment, % fissile atoms. .	7.08	6.75	5.31	3.77
Average core composition :				
Moderator-to-fissile atom ratio	20,400	13,800	13,000	8,700
Enrichment, % fissile atoms. .	3.69	5.08	2.97	3.06
Reactivity lifetime :				
Cycle time, full power days . .	573	349	925	746
Fissions per initial fissionable atom	1.52	0.93	1.72	1.39
Mwd/T (U + Pu + Th) . . .	102,000	59,000	87,000	50,000
Average core specific power, kW/kg fissile	5,160	3,480	3,330	2,230
Net conversion ratio	0.63	0.63	0.72	0.86
Plant throughputs, MT/year :				
Processing	96	172	114	207
Fabrication	108	186	126	219
Unit cost for fabrication plus processing, $/kg	265	197	239	178
Ratio of initial to average power density	1.49	1.21	1.84	1.24
Average η of fissile nuclides	1.92	1.93	2.14	2.19

TABLE IV. — Neutron Balance for Minimum Cost of Homogeneous Fuels

	Reactions Per Source Neutron			
	Partially Enriched Uranium Fuel		Uranium-Thorium Fuel	
	Not Recycled	Recycled	Not Recycled	Recycled
Absorptions :				
Th^{232}			0.334	0.353
Pa^{233}			0.010	0.009
U^{233}			0.228	0.341
U^{234}			0.011	0.043
U^{235}	0.223	0.191	0.235	0.109
U^{236}	0.004	0.029	0.007	0.018
Np^{237}	0.001	0.007	0.003	0.007
U^{238}	0.256	0.235	0.003	0.004
Pu^{239}	0.233	0.235	0.002	0.004
Pu^{240}	0.071	0.092	0.001	0.001
Pu^{241}	0.059	0.090	0.001	0.001
Pu^{242}	0.003	0.023	0.000	0.000
Fission products	0.088	0.055	0.103	0.066
Moderator	0.037	0.022	0.037	0.023
Leakage	0.025	0.021	0.025	0.021
TOTAL	1.000	1.000	1.000	1.000
Productions :				
U^{233}			0.519	0.772
U^{235}	0.448	0.379	0.473	0.216
Pu^{239}	0.411	0.412	0.004	0.006
Pu^{241}	0.132	0.203	0.002	0.003
Other	0.009	0.006	0.002	0.003
TOTAL	1.000	1.000	1.000	1.000

In Table II we have also tabulated the total fuel-cycle cost for non-recycled fuels under the assumption that the spent fuel is to be discarded without reprocessing. The total cost for the partially enriched uranium fuel was decreased to 0.93 mills/kWhr(e) in this case, indicating that it is not worthwhile to reprocess such a feed to recover its plutonium even in a very large-scale reprocessing industry. This somewhat surprising result comes about from a combination of the very high burnup with most of the plutonium burned *in situ* and the high Pu^{242} content of the spent fuel, along with the fact that reprocessing is inherently more expensive for gra-

phite-matrix fuels than for some other types. The cost for the thorium-uranium fuel was increased to 1.01 mills/kwhr(e) by discarding the spent fuel; the economics of this type of fuel evidently requires that the spent fuel be reprocessed and reused.

Estimates of the probable cost of reprocessing the fuel do not depend strongly on the design of the fuel element or the mode of fuel management. Instead, the reprocessing cost depends primarily on the assumptions made regarding the size of the industry and the size of the reactor complex to be served by a single reprocessing plant. This situation is illustrated in Table II where the reprocessing cost varies only from 0.155 to 0.177 mills/kwhr(e) as the type of fuel and the burnup are changed considerably. The situation with regard to fabrication cost is somewhat different. This item can vary significantly with fuel element design, burnup, and type of fuel. There may also be significant changes brought by the degree of automation in the fabrication plants. Hence, it seems worthwhile to consider the sensitivity with which the fuel-cycle cost for each type of fuel varies with the unit cost for fuel fabrication. We have considered the fuel fabrication cost as a parameter to be varied independently and have found the fuel compositions which gave lowest cost for each assumed value. The resulting fuel-cycle costs are plotted in Fig. 6 for three of the types of fuel management : recycled uranium-thorium, non-recycled uranium-thorium with the fuel reprocessed and sold, and partially enriched uranium with the spent fuel discarded. The other possibilities for fuel management (recycled

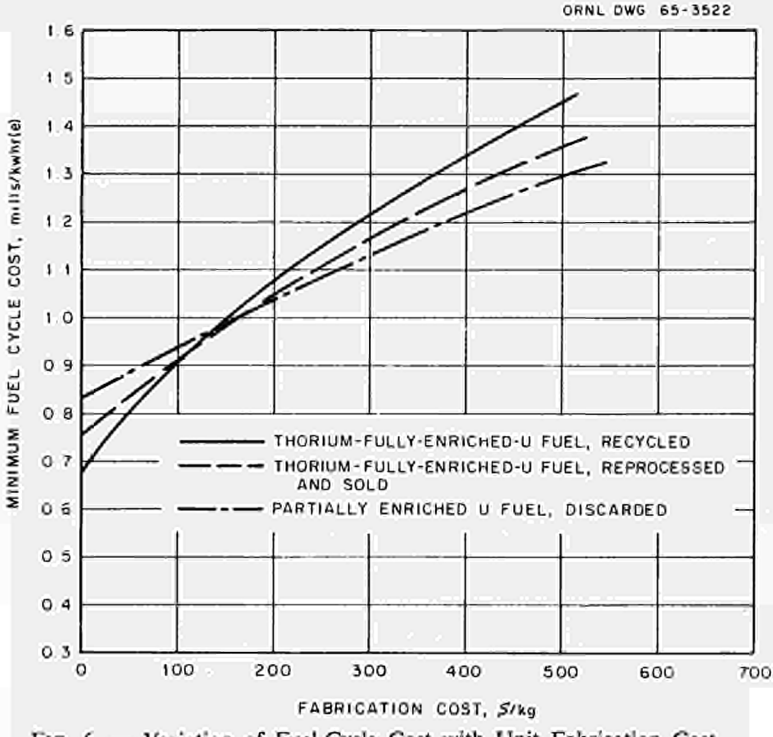

FIG. 6. — Variation of Fuel-Cycle Cost with Unit Fabrication Cost.

partially enriched uranium, non-recycled partially enriched uranium with the fuel reprocessed and sold, and uranium-thorium with the fuel discarded) are not shown because they never gave costs as low as the types shown. The trend in optimization was toward higher enrichments, higher burnups and lower conversion ratios as the fuel fabrication costs was increased.

The data in Fig. 6 indicate that the recycled fully enriched uranium fuel gives a lower fuel-cycle cost than any of the other fuel types when the fabrication cost is below \$100/kg. This fuel is particularly advantageous at low fabrication costs because a low burnup is permitted, thus increasing the conversion ratio. The enhancement in conversion ratio results from the smaller number of neutron captures in fission products and also from a secondary improvement in the η of the fuel when the recycled stream has a higher ratio of U^{233} to U^{235}. At fabrication costs above \$165/kg, the best costs were obtained with partially enriched uranium fuel in which the spent fuel is discarded. At the very high burnups required by these fabrication costs the low fuel inventory cost for the latter fuel is the decisive factor. There is apparently an area of fabrication costs from \$100 to \$165/kg in which the non-recycle uranium-thorium fuel gives the lowest cost. However, such a fuel cycle would imply a guaranteed market for the spent fuel in some other reactor type.

It should be kept in mind that the difference in fuel-cycle cost among the various types of fuel is, in all cases, small in comparison with the total power cost. A more complete analysis would be needed, taking into account any differences in thermal performance and core design requirements, before choosing one type of fuel. Nevertheless, the work we have done indicates particular promise for a partially enriched uranium throwaway cycle for near-term use and for recycled uranium-thorium fuel for ultimate large-scale use.

2. — Heterogeneous Low-Enrichment Uranium Fuel

In considering the use of partially enriched uranium fuels in graphite-moderated high-temperature gas-cooled reactors a question that arises is whether there would be an advantage in "lumping" the fuel. The heterogeneous lattice can achieve a given burnup at a considerably lower enrichment than the homogeneous reactor core and can thereby use U^{235} at a lower cost per gram. The principal offsetting disadvantage is that a smaller volume fraction of the core is used for heat generation when the moderator and fuel are segregated and the power density and specific power are simply limited by heat removal considerations.

We have investigated this question by studying a configuration shown schematically in Fig. 7. The fuel element would consist of a hexagonal block of graphite with a cluster of 42 coolant passages and 19 fuel channels at its center. The fuel channels contained UO_2 at 95 % of theoretical density. The reactor power was 1,000 Mw(e), the core height was 25 *ft*, the coolant (helium) temperature was 720° F at the inlet and 1,470° F at the outlet. The maximum fuel temperature was limited

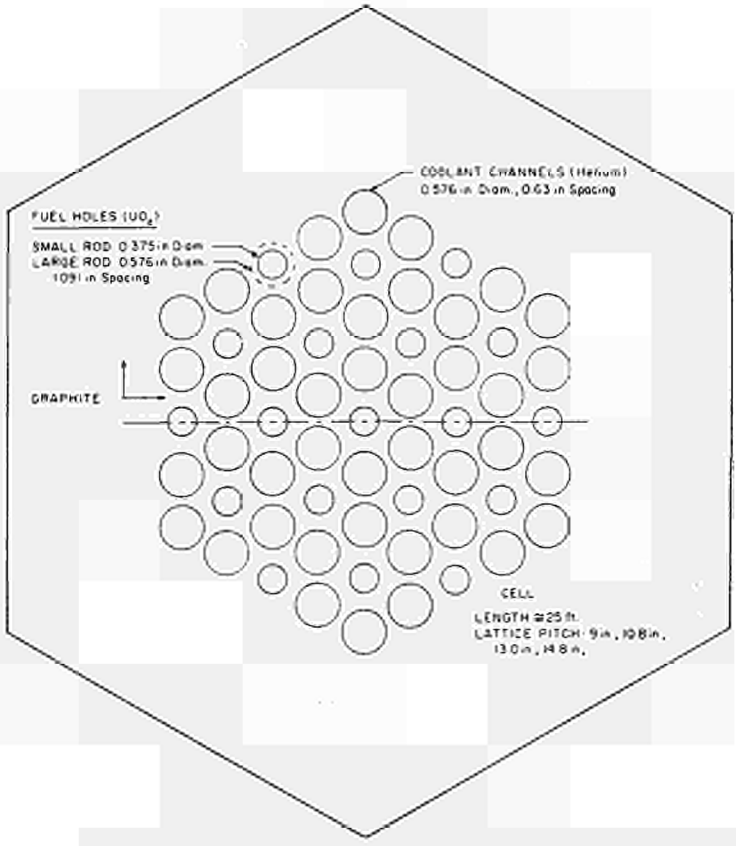

FIG. 7. — Cross Section of Heterogeneous Fuel Element.

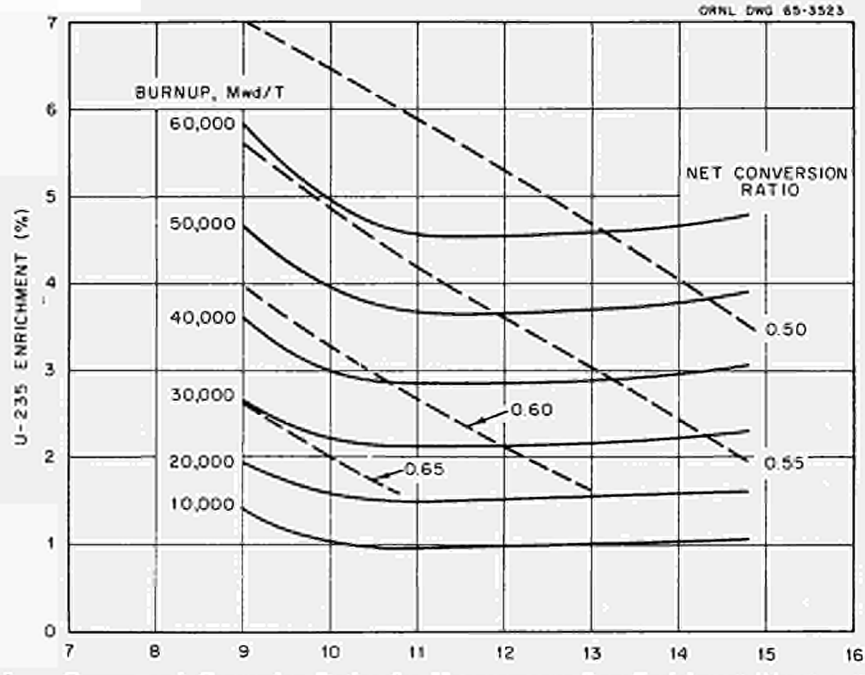

FIG. 8. — Burnup and Conversion Ratios for Heterogeneous Low-Enrichment Uranium Fuel.

to 3032° F, giving a power output of 2.586 Mw for the maximum power fuel element. The fuel was not recycled. Calculational methods were essentially the same as in the study of the homogeneous fuel with heterogeneous resonance integrals computed by the GAM-II [4] code and thermal cells calculated by the THERMOS [5] code. Costs were computed on the same basis as for the homogeneous fuel.

The principal variables were the lattice pitch between fuel clusters, the size of the fuel channel, and the fuel enrichment. The burnups and conversion ratios are shown in Fig. 8 for the 0.375-in.-diam fuel channel. It can be seen that considerably lower enrichments were obtained than in the case of the homogeneous fuel. Criticality was obtained with enrichments as low as 1 %. However, we have not been able to find a configuration which would be critical on natural uranium, largely because the amount of graphite and void space that must be present in the fuel cluster prevent achieving sufficiently small effective resonance integrals for U^{238}. Calculations were also made for a larger rod, of 0.576 in. diam, with very similar results.

The case that gave lowest fuel-cycle costs is listed in Table V. The fuel-cycle cost was computed on the basis of discarding the spent fuel without reprocessing since the lowest costs were obtained in this way. The optimum case had a 2 % feed enrichment in comparison with a 7 % enrichment for the homogeneous non-recycle partially enriched optimum. The total fuel-cycle cost for the heterogeneous case was 0.94 mills/kwhr(e) which was almost identical with the 0.93 mills/kwhr(e) calculated for the homogeneous case. However, the power density of

TABLE V. — Summary of Optimum Case for Heterogeneous Uranium Fuel

Power density, w/cm³	2.58
Initial fuel enrichment, %	2
Lattice pitch, in.	13
Reactivity lifetime :	
Cycle time, full power days	1,400
Fissions per initial fissile atom	1.44
Mwd/T (total U)	27,300
Average specific power, kW/kg fissile	2,220
Net conversion ratio	0.59
Fabrication plant throughput, MT/year	403
Unit cost for fabrication, $/kg	58.5
Fuel-cycle cost, mills/kwhr(e) :	
Fabrication	0.224
Fabrication interest	0.046
Shipping + storage	0.022
Uranium feed	0.561
Core inventory	0.066
Fabrication inventory	0.023
TOTAL	0.942

the heterogeneous case was only 2.58 w/cm^3, implying that the capital costs for the core and pressure vessel would be high. We conclude from these data that it will be difficult to find a configuration for a heterogeneous fuel element which will give, at the same time, very low enrichment and a combination of fuel costs and capital costs which would be attractive.

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URANIUM 235 FUELED THERMAL HIGH TEMPERATURE REACTOR WITH URANIUM 233 IN EQUILIBRIUM STATE WITH THORIUM 232 AS THE FERTILE SPECIES

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ABSTRACT

This paper gives the description and the results of calculations for favourable operating conditions of an uranium 235 fueled, graphite-moderated, helium cooled high temperature reactor with thorium 232 as fertile material supposing equilibrium state of uranium 233 in thorium at every time (THTR-Project). The conversion factors and special defined fifa-values are calculated on the basis of present nuclear cross-section data.

1. — INTRODUCTION

A thermal thorium converter reactor utilizing uranium-235 as fuel and thorium-232 as fertile nuclide is studied by assuming equilibrium condition of uranium-233 in the fertile material. A high temperature helium cooled graphite moderated reactor of the type of a pebble bed reactor is proposed. This reactor concept combines the advantages of low neutron absorption in graphite, the absence of absorbing structural materials and small leakage rates with the possibility to separate fuel and fertile material thus getting different lifetimes for both kinds of fuel elements in the reactor core.

The uranium-235 is used in high enrichment in order to prevent the build up of plutonium, which makes worse the conversion rate of Th^{232}-U^{233}. The low portion of uranium-238 is neglected in the calculations.

The charging of the reactor is done continuously. Consequently the excess reactivity, compensated by control rods, is not necessary. To maintain the critical condition the reactor is fed with fresh fuel and thorium balls (1), at the same time the balls with the highest burnup are removed. In doing so the volume of the core is kept constant.

To burnup all the balls alike — which is assumed in these calculations — they are rolled round continuously. Theoretically this equal burnup of all the balls will be obtained by rolling them round with infinite speed. Practically this state can already be obtained with finite speed.

(1) Fuel balls : graphite balls containing coated particles of U^{235} in the core; thorium balls : graphite balls containing coated particles of thorium dioxide in the core.

Because the equilibrium status will not be reached during the first time of operation, this phase shall be disregarded. After having obtained stationary conditions, there are the same number of balls of every type in every burnup state in the core. That means that the temporal average of the progress of the concentrations of all the nuclides in one ball of every type delivers the steady-state concentrations of all the fissile and fertile nuclides and fission products in the core. In this model the fissionable and fertile material are treated as if intimately mixed and irradiated in a single reactor region.

2. — THEORY

2.1. — GENERAL.

The lifetime T_2 of the balls containing the fertile material is introduced as parameter. Then a definite lifetime T_1 of the fuel balls containing U^{235} fulfills the criticality equation for a given set of neutron temperature, average power density, neutron leakage, moderator-to-uranium (1) ratio, and moderator-to-thorium ratio.

For that purpose a reaction rate for absorption may be defined in the following way :

$$s = \int_0^{x_e} \sigma_a(x) \varnothing(x) dx \; ; \left[\frac{1}{a}\right]$$

with $x = \sqrt{\frac{E}{kT_N}}$;

x_e = corresponding $E = 10$ MeV;
E = energy of neutron (eV);
k = Boltzmann constant (eV. $^oC^{-1}$);
T_N = neutron temperature (^oK);
$\sigma_a(x)$ = microscopic cross-section for absorption in dependence of $x(cm^2)$;
$\varnothing(x)$ = neutron flux in dependence of $x(cm^{-2}a^{-1})$.

By this the conversion factor C is given by

$$C = \frac{s_2 N_2}{s_2 N_2 + s_1 N_1}$$

assuming equilibrium state of U^{233} in Th^{232},

with N_i = atomic concentration of material $i(cm^{-3})$;
index 1 = U^{235};
index 2 = U^{233}.

To get an idea of the best utilization of uranium-235 unlike the usual convention a fifa-value is defined :

$$\text{fifa} = \frac{T_1(s_1^f N_1 + s_2^f N_2)}{N_{10}} = \text{number of fissions per initial invested } U^{235} \text{ atom,}$$

(1) " Uranium " consists of the mean concentrations of U^{235} and U^{233}.

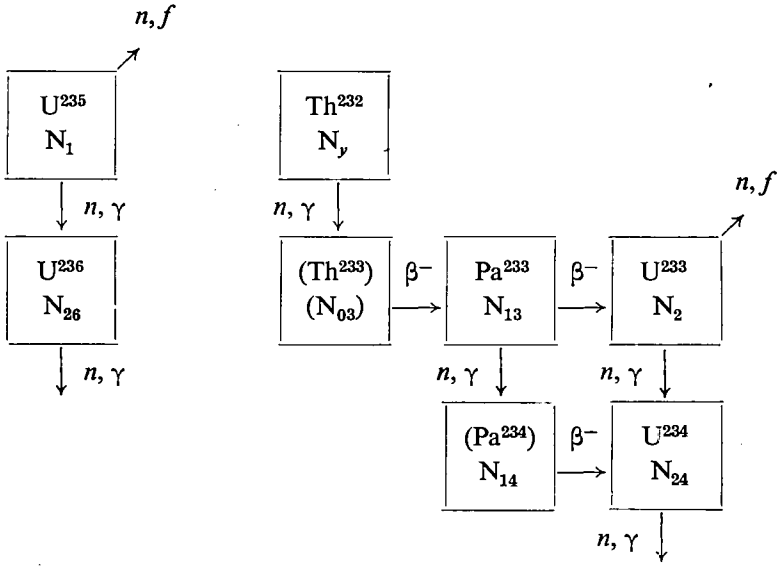
with s^f = reaction rate for fission;
N_{10} = initial atomic concentration of U^{235}.

2.2. — Burnup Equations.

The irradiation chains of U^{235} and Th^{232} are given in Table 1. Concentrations shown enclosed in parentheses have been treated as if negligibly small because of the short half-lives of those elements. The chains are terminated by the absorption of a neutron by U^{236}, U^{234} respectively. That is allowed by taking into consideration the error of the known neutron cross-section data.

The table illustrates the notation.

TABLE 1. — Irradiation Scheme of U^{235} and Th^{232}



For the description of the change in the atomic concentrations the following differential equations can be written. Simultaneously the mean concentrations are given :

U^{235} :

$$\frac{dN_1}{dt} = -s_1 N_1 \; ;$$

$$\bar{N}_1 = \frac{N_{10}}{s_1 T_1} (1 - e^{-s_1 T_1}) \; ;$$

U^{236} :

$$\frac{dN_{26}}{dt} = \frac{\alpha_1}{1 + \alpha_1} s_1 N_1 - s_{26} N_{26} \; ;$$

$$N_{26} = \frac{1}{s_{26}} \frac{\alpha_1}{1 + \alpha_1} \frac{s_1 \bar{N}_1}{(s_1 - s_{26})} \left[\frac{1 - e^{-s_{26} T_1}}{1 - e^{-s_1 T_1}} s_1 - s_6 \right] ;$$

Th^{232} :

$$\frac{dN_y}{dt} = 0 \; ; \qquad \text{(assumption)}$$

$$N_y = \text{const} \; ;$$

Pa^{233} :

$$\frac{dN_{13}}{dt} = s_y N_y - (\lambda_{13} + s_{13}) N_{13} \; ;$$

$$\bar{N}_{13} = \frac{s_y N_y}{\lambda_{13} + s_{13}} \text{ (equilibrium status of } U^{233} \text{ in } Th^{232});$$

U^{233} :

$$\frac{dN_2}{dt} = \lambda_{13} N_{13} - s_2 N_2 \; ;$$

$$\bar{N}_2 = \frac{\lambda_{13}}{s_2} \frac{s_y N_y}{\lambda_{13} + s_{13}}, \text{ (equilibrium status in } Th^{232});$$

Pa^{234} :

$$\frac{dN_{14}}{dt} = s_{13} N_{13} - (\lambda_{14} + s_{14}) N_{14} \; ;$$

$$\bar{N}_{14} = \frac{s_{13} N_{13}}{\lambda_{14}}, \; (\lambda_{14} \gg s_{14}, \text{ short half-life});$$

U^{234} :

$$\frac{dN_{24}}{dt} = \frac{\alpha_2}{1 + \alpha_2} s_2 N_2 + \lambda_{14} N_{14} - s_{24} N_{24} \; ;$$

$$\bar{N}_{24} = \frac{\alpha_2 \lambda_{13} + (1 + \alpha_2) s_{13}}{(1 + \alpha_2)(\lambda_{13} + s_{13})} \frac{s_y N_y}{s_{24}} \left[1 - \frac{1}{s_{24} T_2} (1 - e^{-s_{24} T_2}) \right] ;$$

with λ = decay constant $(1/a)$;

$\alpha = \frac{s^c}{s^f}$ = capture-to-fission ratio.

For the study of the change in the composition of the various fission products three groups are distinguished :

1. stable fission products,
2. decaying fission products,
3. stable absorbing nuclides following decaying fission products.

Only fission products with an absorption cross-section of at least more than 5 barns have been considered particularly. The remainder has been condensed to one stable pseudo fission product. Table 2 shows the fission products.

TABLE 2. — Fission Products

Group	
1	Kr^{83}, Tc^{99}, Pd^{107}, Pd^{108}, Ag^{109}, Cd^{113}, In^{115}, I^{127}, I^{129}, Xe^{131}, Cs^{133}, Cs^{135}, La^{139}, Nd^{145}, Nd^{146}, Sm^{149}, Sm^{151}, Sm^{152}, Eu^{153}, Sm^{154}, Gd^{157}
2	Kr^{85}, Rh^{105}, Xe^{133}, Xe^{135}, Pr^{143}, Pm^{147}, Eu^{155}
3	Mo^{95}, Rh^{103}, Pd^{105}, Pr^{141}, Nd^{143}, Nd^{144}, Sm^{147}

The differential equations and the mean concentrations of the fission products of the various groups are determined by the following formulae :

For fission products of U^{235}

group 1 :

$$\frac{dN_i^{(1)}}{dt} = \frac{\gamma_i^{(1)}}{1+\alpha_1} s_1 N_1 - s_i N_i^{(1)} ;$$

$$\overline{N}_i^{(1)} = \frac{1}{s_i} \frac{\gamma_i^{(1)}}{1+\alpha_1} \frac{s_1 \overline{N}_1}{(s_1 - s_i)} \left[\frac{1 - e^{-s_i T_1}}{1 - e^{-s_1 T_1}} s_1 - s_i \right] ;$$

group 2 :

$$\frac{dN_i^{(1)}}{dt} = \frac{\gamma_i^{(1)}}{1+\alpha_1} s_1 N_1 - (\lambda_i + s_i) N_i^{(1)} ;$$

$$\overline{N}_i^{(1)} = \frac{1}{s_i} \frac{\gamma_i^{(1)}}{1+\alpha_1} \frac{s_i}{\lambda_i + s_i - s_1} s_1 \overline{N}_1 \left[1 - \frac{s_1}{\lambda_i + s_i} \frac{1 - e^{-T_1(\lambda_i + s_i)}}{1 - e^{-s_1 T_1}} \right]$$

group 3 :

$$\frac{dN_j^{(1)}}{dt} = \lambda_i N_i^{(1)} - s_j N_j^{(1)} ;$$

$$\overline{N}_j^{(1)} = \frac{1}{s_j} \frac{\gamma_i^{(1)}}{1+\alpha_1} \frac{s_j}{\lambda_i + s_i - s_1} \frac{\lambda_i}{s_j - s_1} s_1 \overline{N}_1 \left\{ 1 - \frac{s_1}{(\lambda_i + s_i - s_j)(1 - e^{-s_1 T_1})} \right.$$
$$\left. \left[\frac{\lambda_i + s_i - s_1}{s_j} (1 - e^{-s_j T_1}) - \frac{s_j - s_1}{\lambda_i + s_i} (1 - e^{-T_1(\lambda_i + s_i)}) \right] \right\} ;$$

for fission products of U^{233}

group 1 :

$$\frac{dN_i^{(2)}}{dt} = \frac{\gamma_i^{(2)}}{1+\alpha_2} s_2 N_2 - s_i N_i^{(2)} ;$$

$$\overline{N}_i^{(2)} = \frac{1}{s_i} \frac{\gamma_i^{(2)}}{1+\alpha_2} s_2 \overline{N}_2 \left[1 - \frac{1 - e^{-s_i T_2}}{s_i T_2} \right] ;$$

group 2 :

$$\frac{dN_i^{(2)}}{dt} = \frac{\gamma_i^{(2)}}{1+\alpha_2} s_2 N_2 - (\lambda_i + s_i) N_i ;$$

$$\overline{N}_i^{(2)} = \frac{1}{s_i} \frac{\gamma_i^{(2)}}{1+\alpha_2} \frac{s_i}{\lambda_i + s_i} s_2 N_2 \left[1 - \frac{1 - e^{-T_2(\lambda_i + s_i)}}{T_2(\lambda_i + s_i)} \right] ;$$

group 3 :

$$\frac{dN_j^{(2)}}{dt} = \lambda_i N_i^{(2)} - s_j N_j^{(2)} ;$$

$$\overline{N}_j^{(2)} = \frac{1}{s_j} \frac{\gamma_i^{(2)}}{1+\alpha_2} \frac{\lambda_i}{\lambda_i + s_i} s_2 \overline{N}_2 \left\{ 1 - \frac{\lambda_i + s_i}{T_2 s_j (\lambda_i + s_i - s_j)} \left[(1 - e^{-s_j T_2}) \right. \right.$$
$$\left. \left. - \frac{s_j^2 (1 - e^{-T_2(\lambda_i + s_i)})}{(\lambda_i + s_i)^2} \right] \right\} ;$$

with γ_i = fission yield of forming a nuclide i.

2.3. — Calculation of Criticality.

The criticality condition for a homogeneous reactor can be written according to the four-factor formula :

$$(\nu_1 s_1{}^f\bar{N}_1 + \nu_2 s_2{}^f\bar{N}_2)\Gamma_s = s_1\bar{N}_1 + s_2\bar{N}_2 + s_y\bar{N}_y + s_{13}\bar{N}_{13} + s_{24}\bar{N}_{24} + s_{26}\bar{N}_{26} + \Sigma s_i\bar{N}_i{}^{(1)} + \Sigma s_i\bar{N}_i{}^{(2)} + s_M N_M(1 + L_M{}^2B^2)$$

with Γ_s = nonleakage probability while slowing down;
ν = number of neutrons emitted in fission;
L_M = diffusion length of the moderator (cm);
B^2 = buckling of the system (cm^{-2});
index M = referred to moderator.

Assuming equilibrium state for U^{233} in Th^{232} and introducing the quantity $\eta = \frac{\nu_s{}^f}{s}$, the equation above takes the form :

$$s_1\bar{N}_1(\eta_1\Gamma_s - 1) + s_2\bar{N}_2\left(\eta_2\Gamma_s - 2(1 + \frac{s_{13}}{\lambda_{13}})\right) = s_{24}\bar{N}_{24} + s_{26}\bar{N}_{26} + \Sigma s_i N_i{}^{(1)} + \Sigma s_i\bar{N}_i{}^{(2)} + s_M N_M(1 + L_M{}^2B^2).$$

The terms on the right side of this equation are given in section 2.2. They are proportional to $s_1\bar{N}_1$, $s_2\bar{N}_2$ respectively.

When the lifetime T_2 is given, the lifetime T_1 can be obtained from the transcendental equation above by iteration procedure for fixed values of $\bar{N}_1$, $\bar{N}_2$ N_M, and Γ_s.

The buckling B^2 is calculated from

$$B^2 = \frac{1 - \Gamma_s}{\Gamma_s\tau_{th}} \text{ with } \tau_{th} = \text{Fermi age } (cm^2).$$

2.4. — Spectrum Calculation.

The reaction rate s is divided up into a thermal and an epithermal portion

$$s = \int_0^{x_{th}} \sigma(x)\varnothing(x)dx + \int_{x_{th}}^{x_e} \sigma(x)\varnothing(x)dx = s_{th} + s_{epi}.$$

In order to obtain reaction rates in the thermal region, the thermal neutron flux must be determined. Because of the high neutron temperature of about 900° K in a high temperature reactor, the crystalline effects in graphite may be neglected in the first approximation. The thermal neutron flux spectrum will now be calculated with the aid of the Wilkins equation [3] :

$$x\frac{d^2n}{dx^2} + (2x^2 - 1)\frac{dn}{dx} + (4x - \Delta(x))n = 0 \text{ (Heavy gas model)}$$

with n = neutron density (cm^{-3});

$$\Delta(x) = \frac{4\Sigma^a(x)x}{\frac{2}{A}\Sigma^s(x)};$$

Σ^a = total homogenized macroscopic absorption cross-section (cm^{-1});

Σ^s = total homogenized macroscopic scattering cross-section (cm^{-1});

A = averaged atomic weight.

For numerical solution the differential equation is formed by substitution of

$$y_1 = n(x),$$

$$y_2 = x\frac{dn}{dx}$$

into a system of two differential equations of first order

$$\frac{dy_1}{dx} = \frac{y_2}{x},$$

$$\frac{dy_2}{dx} = (2 - 2x^2)\frac{y_2}{x} + (\Delta(x) - 4x)y_1.$$

The numerical integration is done with the Runge-Kutta method. The neutron flux follows from

$$\varnothing(x) = n(x)x\sqrt{\frac{2kT_N}{m_N}} \text{ with } m_N = \text{rest mass of the neutron } (eVs^2\ cm^{-2}).$$

The epithermal region of the neutron flux spectrum is determined by solving 22-groups diffusion equations for infinite medium. Because of the large dimensions of the proposed reactor, the nonleakage probability while slowing down is supposed to be not smaller than 0.98. Then the influence of the leakage on the spectrum may be neglected. The neutron energy range is extended from 0.683 eV to 10 MeV.

The diffusion equation is written in the following form :

$$-\Sigma_a{}^n\varnothing^n - \Sigma_r{}^{n\to n+1}\varnothing^n + \Sigma^{n-1\to n}\varnothing^{n-1} + x^n = 0$$

with $\varnothing^n$ = neutron flux in group n;

$\Sigma_r{}^{n\to n+1}$ = macroscopic cross-section for neutrons removed to the next lowest energy group by scattering;

x^n = number of neutrons per fission going in group n.

It follows

$$\varnothing^n = \frac{\Sigma_r{}^{n-1\to n}\varnothing^{n-1} + x^n}{\Sigma_a{}^n + \Sigma_r{}^{n\to n+1}}$$

with $\varnothing^o = O$.

The group lethargy interval was chosen to be 0.75, then the neutrons are removed practically only to the neighbouring energy groups when using graphite as moderator.

After solving the diffusion equations for all the groups, substitution in x is made :

$$\varnothing^n(x) = \varnothing^n(u) \frac{\Delta u^n}{\Delta x^n} \text{ with } u = \text{lethargy.}$$

Now the thermal and epithermal spectrum must be combined by normalizing both parts of the spectrum to the group flux in the range 0.683 — 1.44 eV.

The reaction rates due to normalized spectrum are obtained by integrating the energy dependent cross-sections over the neutron flux using Simpson's rule in case of Th^{232}, U^{233}, U^{235}, Pa^{233}. The reaction rates of the other elements are calculated with the aid of the trapezoidal rule in order to save time.

In order to get the actual reaction rates the load dependent factor ω must be determined :

$$\omega(s_1^{f*}\bar{N}_1 + s_2^{f*}\bar{N}_2) = \frac{Q}{\varepsilon}$$

with $\omega\ s^* = s$ = reaction rate;
s^* = reaction rate due to normalized spectrum;
Q = average power density (MW/m^3);
ε = energy release per fission.

3. — Lay-out of the Programme

In the following a short description of the programme is given.

The input cross-section data have been taken from the references [6], [7], and [8]. They have been arranged for the purpose of these calculations. The integration of the thermal range of the spectrum is done in 30 steps up to 0.683 eV and in other 6 steps up to 1.44 eV to get the range to normalize the spectrum. For the calculations the neutron temperature of 900° K is fixed. Further data for thermal cross-sections, decay constants, and fission yields are taken from reference [4].

The programme can be handled in two ways. For a rapid survey the calculations can be done by taking into consideration for the spectrum calculation only the elements Th^{232}, U^{233}, U^{235} and graphite. The second calculation method allows the determination of the flux spectrum using all the heavy elements and fission products.

A general flow scheme of the programme is shown in Figure 1. First the average power density is given to 7 MW/m^3. In doing so a large reactor of more than 1,000 MW_{el} is assumed with fast leakage of less than 2 %. Then the moderator-to-uranium ratio and the moderator-to-thorium ratio is chosen. After estimating the equilibrium state of U^{233} in Th^{232}, the spectrum is calculated, the reaction rates for those heavy elements, which are used to calculate the equilibrium status, are determined, and the equilibrium status is checked. A given accuracy is reached after some iterations. Then the reaction rates of all the fission products can be calculated. With lifetime T_2, which is corresponding to a given burnup state of the thorium balls the critical condition is solved for lifetime T_1 by iteration procedure with Newton's method and for small values of T_1 with regula falsi.

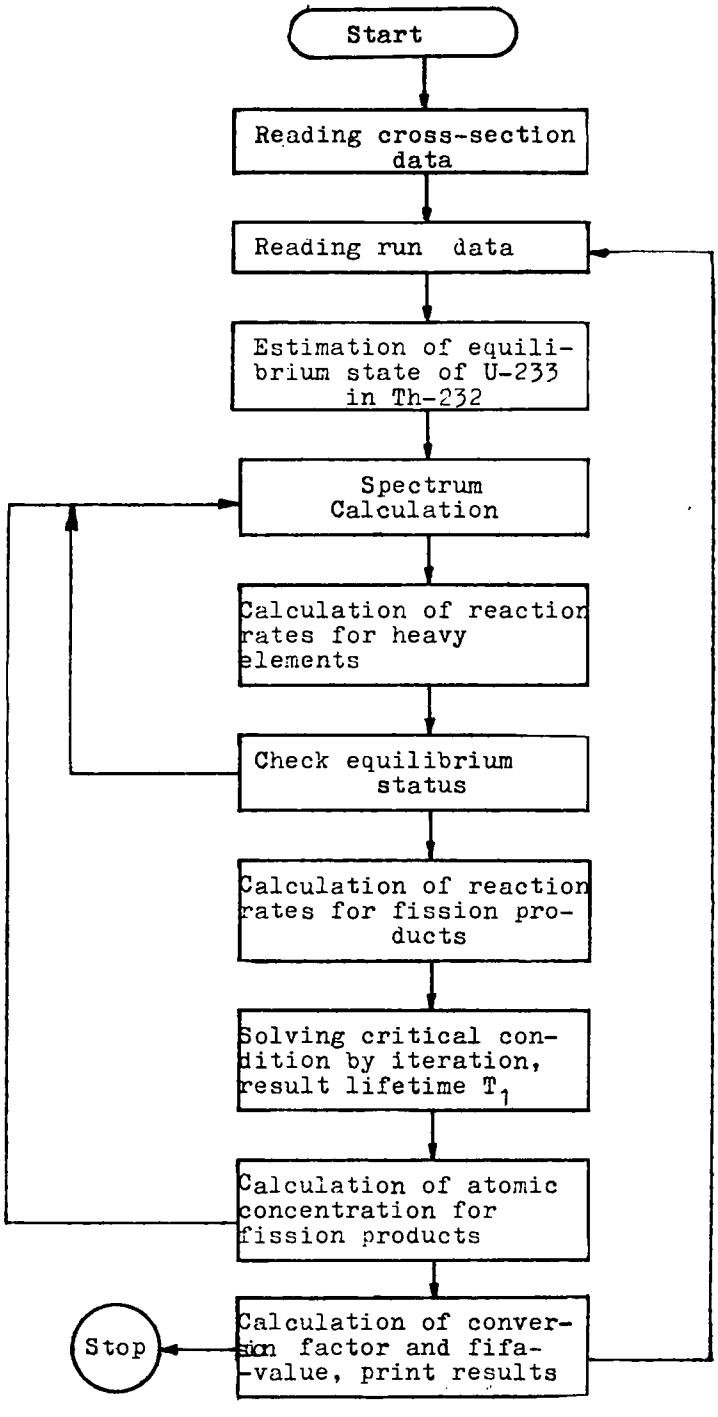


FIG. 1. — Flow Diagram.

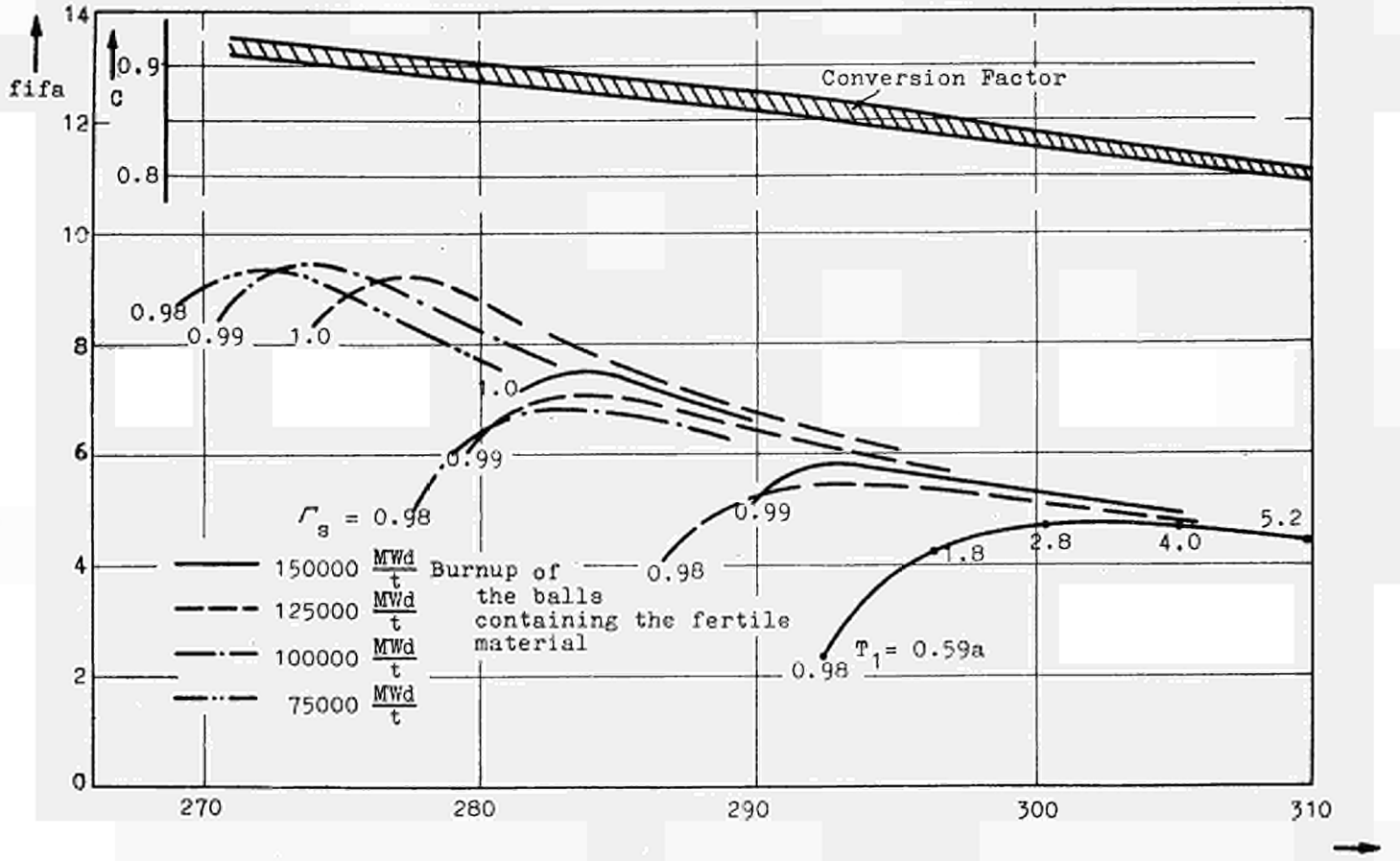


FIG. 2. — Fifa-values and Conversion Factors for $\frac{N_{moder.}}{N_{uranium}} = 6000$, $\frac{N_{moderator}}{N_{thorium}} \rightarrow T = 900^{o}$ K, $Q = 7 \frac{MW}{m^3}$

(Fission Products used in Spectrum Calculation).

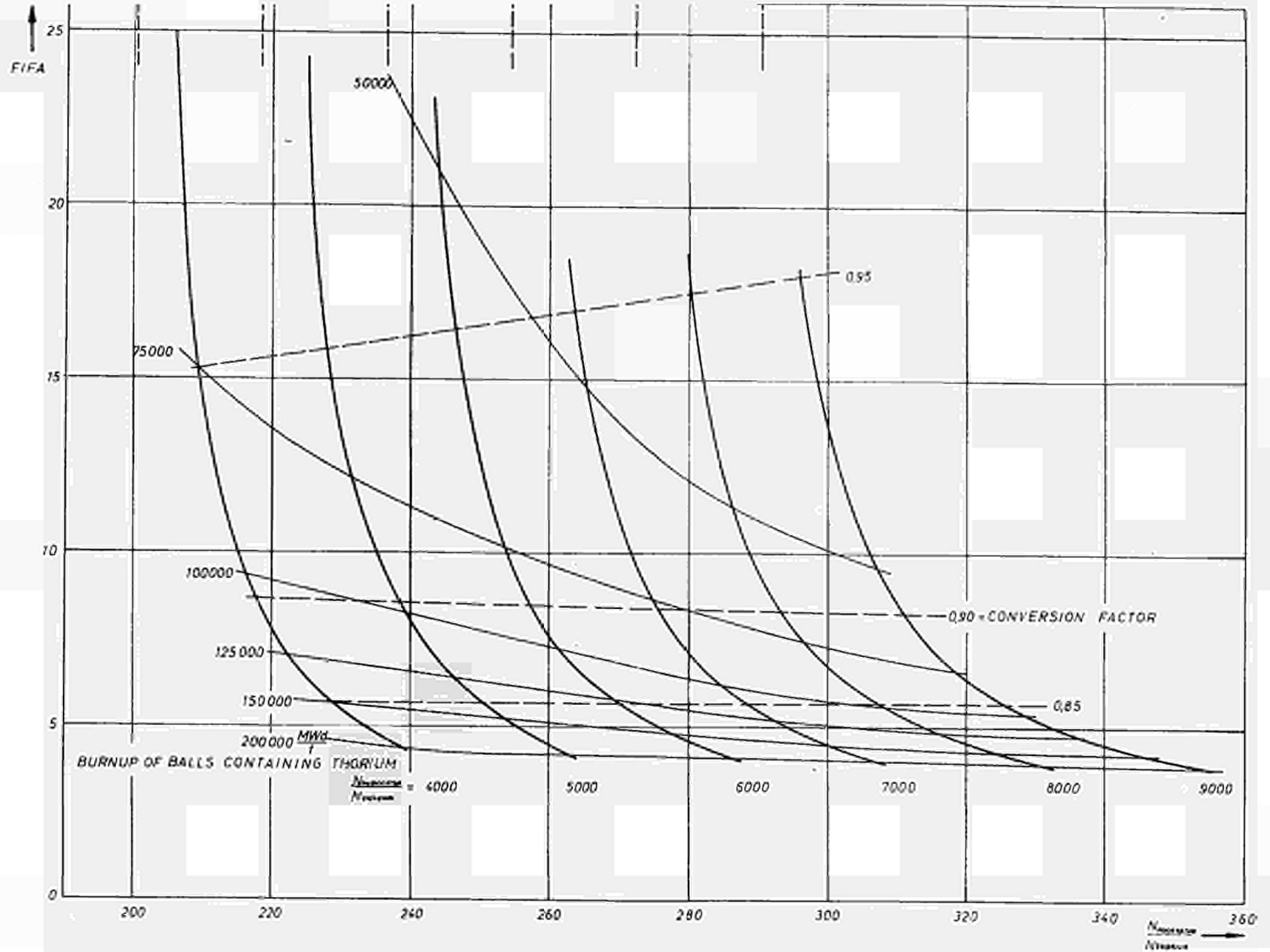


FIG. 3. — Optimum Fifa-values. T = 900° K. Q = 7 MW/m³. Ts = 0.98 (Fission Products not used in Spectrum Calculation)

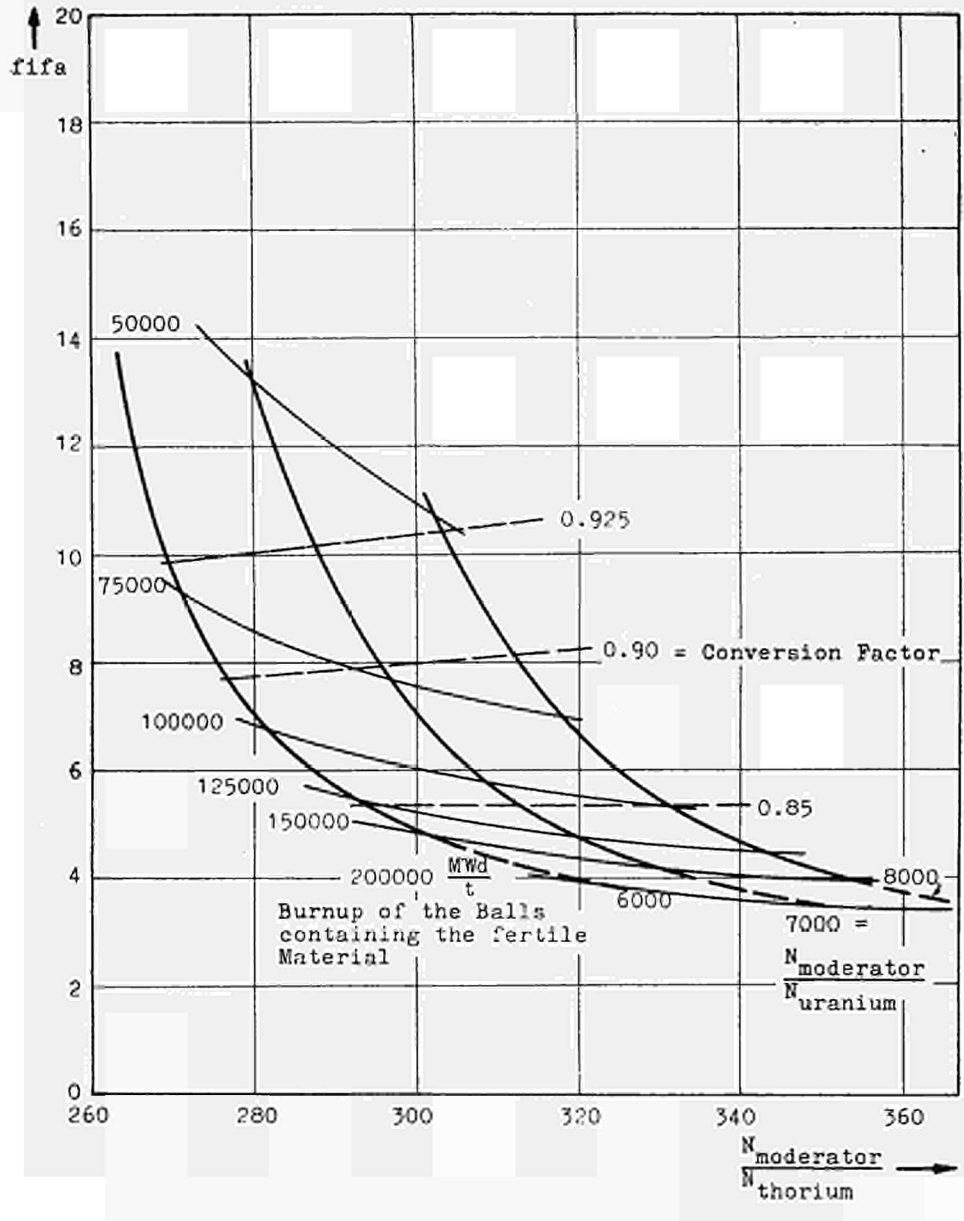


FIG. 4. — Optimum Fifa-values $T = 900°$ K, $Q = 7 \frac{MW}{m^3}$, $\Gamma s = 0.98$

(Fission Products used in Spectrum Calculation.)

In case of consideration the fission products in the neutron flux spectrum the mean atomic concentrations of these nuclides are calculated and again the programme is started with the spectrum calculation. This cycle is repeated until a specified

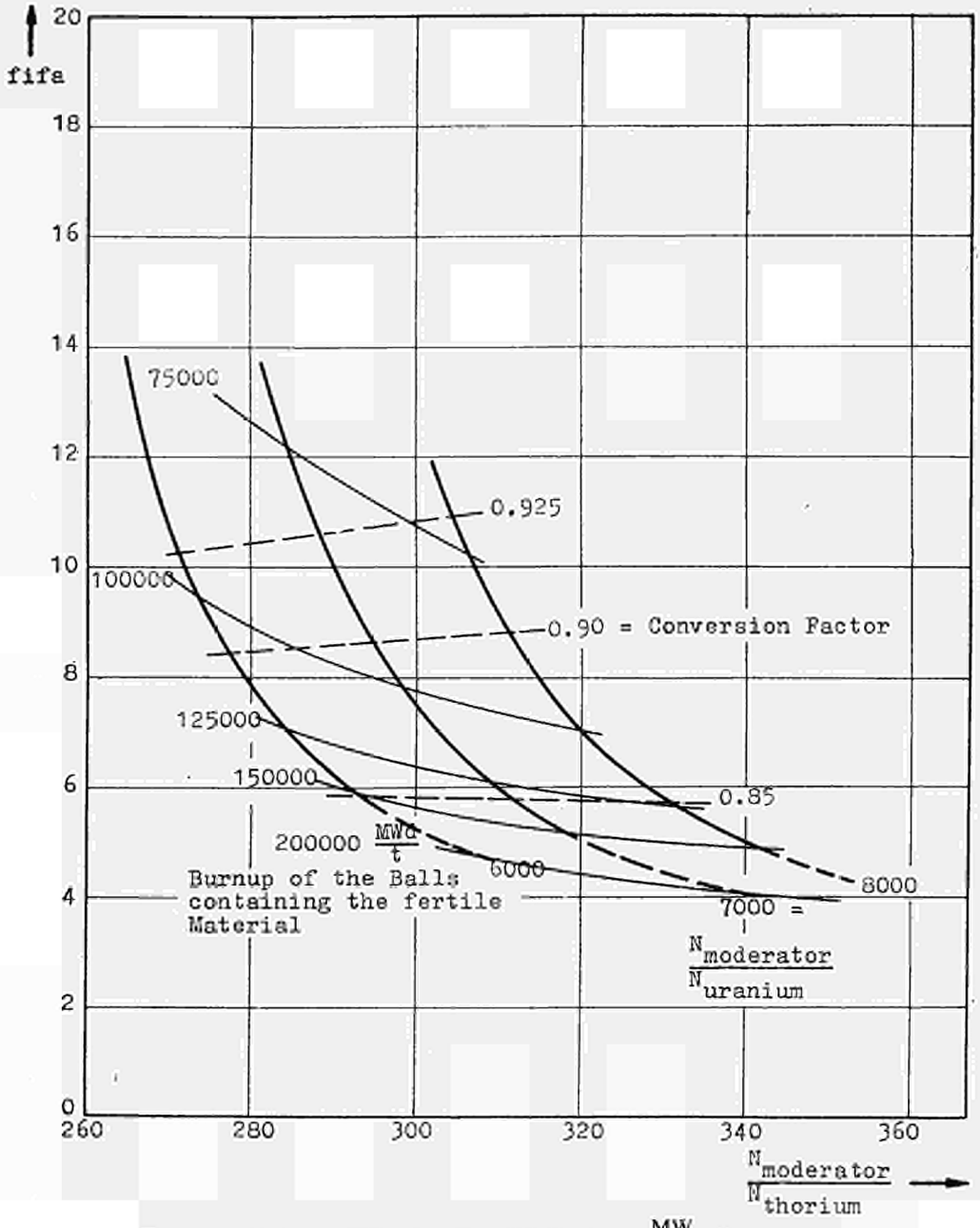


FIG. 5. — Optimum Fifa-values T = 900° K, $Q = 7 \frac{MW}{m^2}$, I's = 0.99.

(Fission Products used in Spectrum Calculation.)

accuracy in the calculation of the equilibrium state has been reached. The conversion factor and the fifa-value will be evaluated and printed, and the whole procedure will be repeated with new input data.

The whole work was done on the Sie 2002 computer of the Rechenzentrum of the Technische Hochschule Aachen.

4. — Results

Typical results are shown in Figure 2. For a given moderator-to-uranium ratio the burnups of the thorium balls and the leakages are varied. Here optimum fifa-values corresponding to a definite burnup of the fuel balls can be found. By keeping constant the moderator-to uranium ratio and the leakage, the optimum fifa-values can be represented in a curve (Figure 3, 4, 5). For a definite moderator-to-uranium ratio the optimum burnup of the fuel balls (i.e. lifetime T_1) and the enrichment of U^{233} in Th^{232} corresponding to the equilibrium status are independent to a certain extent of the burnup of the thorium balls and the leakage. They are only a function of the moderator-to-uranium ratio (Figure 6 and 7). Regarding the representation of the optimum fifa-values in Fig. 3, 4 and 5, it is seen that for conversion factors $C = 1$ the curves run to infinity according to the definition of fifa. With increasing moderator-to-uranium ratio the neutron absorption in Pa^{233} leads to lower fifa for the same burnups of the thorium balls due to higher specific power density. By keeping constant the average power density of 7 MW/m^3 the specific power density Q_s is dependent on the moderator-to-uranium ratio. Values are shown in Table 3.

The burnups of the thorium balls correspond to definite lifetimes T_2, which are useful to know for later cost calculations. They are shown in Figure 8.

To evaluate the favourable conditions for reactor operation an uniform distribution of power density is assumed. This demands the same amount of fissionable material in both kinds of fuel elements. Because all the balls are of the same design, there are five times more thorium balls necessary than fuel balls to fulfill the condition above. Because of the restricted capacity of the balls the moderator-to-thorium ratios are limited to values higher than 300. For this reason only moderator-to-uranium ratios higher than 6,000 are of interest.

To gain a more detailed description for the best operating conditions some calculations of costs must be done.

For making a rough guess for the costs of the reactor core a price of 45 DM/g U^{235} and 100 DM/kg ThO_2 is assumed. The fabrication costs shall consist of the costs for reprocessing and fabricating the coated particles, the fabrication costs of the graphite balls, and the costs for graphite.

Per MW_{th} power the investigated reactor needs the following amounts P of fuel and fertile material yearly :

$$\text{Fuel } P_1 = \frac{N_{10} A_{235}}{T_1 L o Q} \left[\frac{t}{MW_{th} \cdot a}\right]$$

$$\text{Fertile Material } (ThO_2) \int P_2 = \frac{(N_y + N_2) A_{264}}{T_2 L o Q} \left[\frac{t}{MW_{th} \cdot a}\right]$$

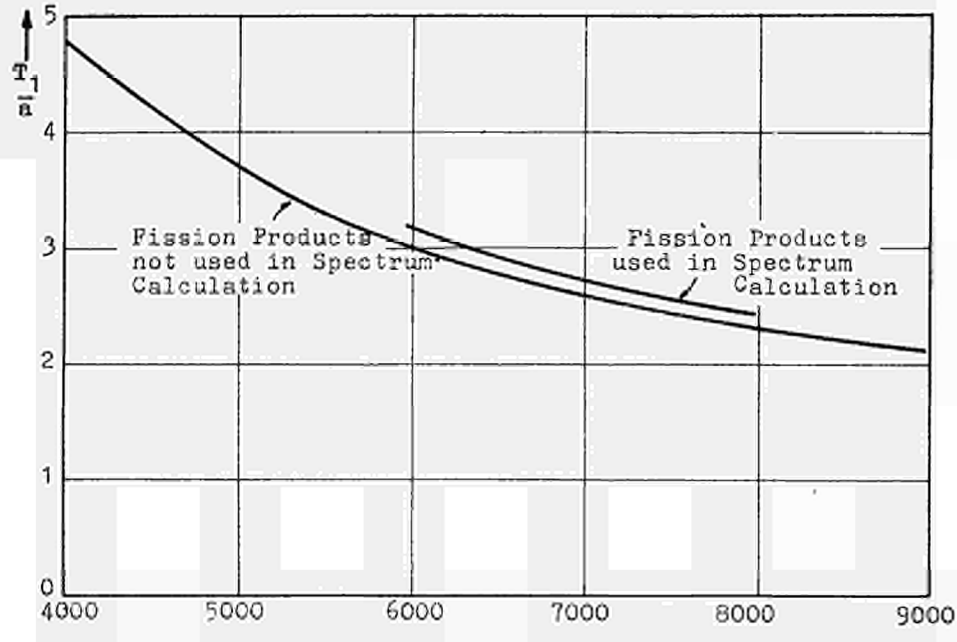


FIG. 6. — Lifetime T_1 for optimum Fifa-values (Fig. 3 and 4).

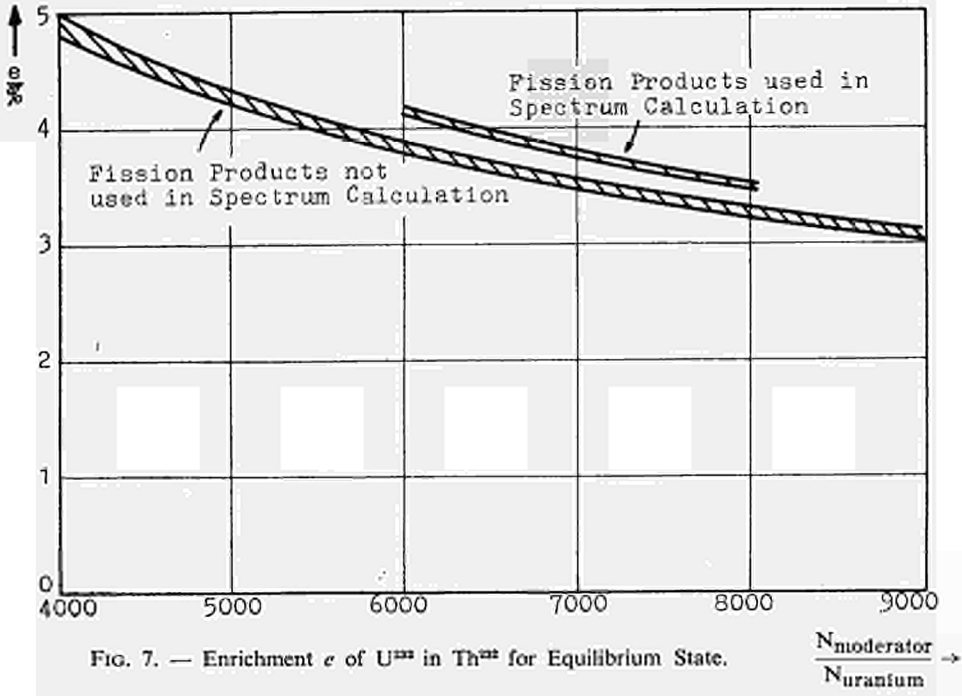


FIG. 7. — Enrichment ε of U^{233} in Th^{232} for Equilibrium State.

TABLE 3. — Specific Power Density

$\frac{N_{moderator}}{N_{uranium}}$	Q_s [kW/g]
4,000	1.41
5,000	1.76
6,000	2.11
7,000	2.46
8,000	2.82
9,000	3.17

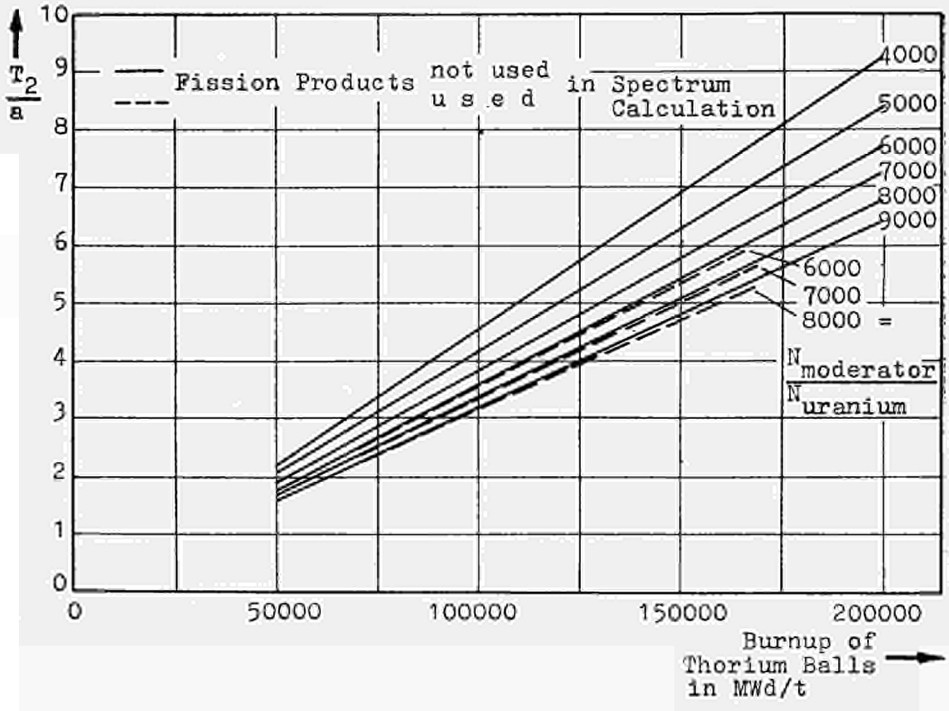


FIG. 8. — Lifetime T_2 corresponding to the Burnup of the Balls containing Thorium (for Fig. 3, 4 and 5).

with Lo = Loschmidt's number,

A = atomic weight.

After having introduced fifa and N_M/N_y, these terms may be written

$$P_1 = \frac{A_{235}}{\text{fifa} \cdot \varepsilon \cdot Lo},$$

$$P_2 = \frac{A_{264} N_M}{T_2 Lo Q N_M/N_y} \left(1 + \frac{N_2}{N_y}\right).$$

If the data of the known quantities are put in the formulae above, the result is :

$$P_1 = \frac{4.4 \cdot 10^{-5}}{\text{fifa}} \left[\frac{\vartheta U^{235}}{kWh_{th}}\right],$$

$$P_2 = \frac{0.366}{T_2[a]N_M/N_y}\left(1 + \frac{N_2}{N_y}\right)\left[\frac{g\text{fertile mat.}}{kWh_{th}}\right].$$

Since the prices for U^{235} and ThO_2 and the fabrication costs are known, the core costs can be calculated.

These costs are shown in Figure 9. The fabrication costs are varied. It is seen that with decreasing fabrication costs the minimum core costs are moving to lower burnups of the balls containing thorium with uranium-233 in equilibrium state, i.e. to higher conversion factors.

After all the fabrication costs are decisively important for the best design of this reactor.

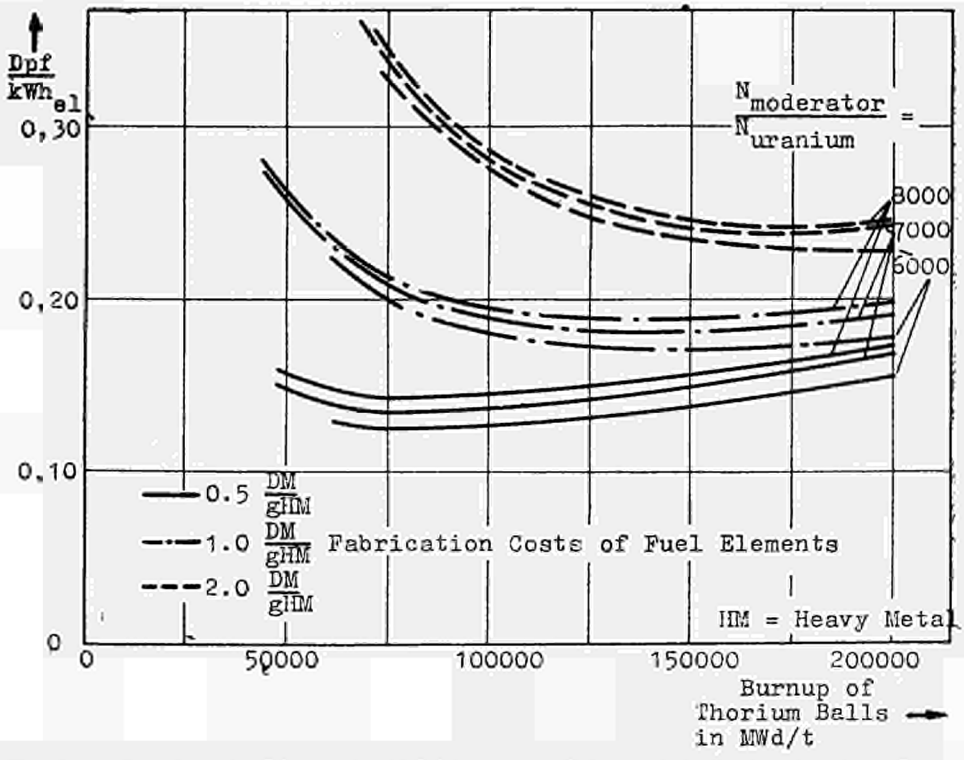


FIG. 9. — Core Costs (45 DM/$g_{U^{235}}$, 100 DM/kg_{ThO_2}, efficiency 40 %, further data from Fig. 4).

ACKNOWLEDGMENT

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FUEL CYCLES OF THE « THORIUM-HOCH-TEMPERATUR-REAKTOR » THTR

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ABSTRACT

In the present paper equilibrium fuel cycles of the Thorium/Uranium/Graphite system are studied. The fuel cycle costs are estimated for different specific fabrication and reprocessing costs.

1. — INTRODUCTION

The main purpose of the present paper is to discuss some results of THTR fuel cycle studies with respect to various economic aspects.

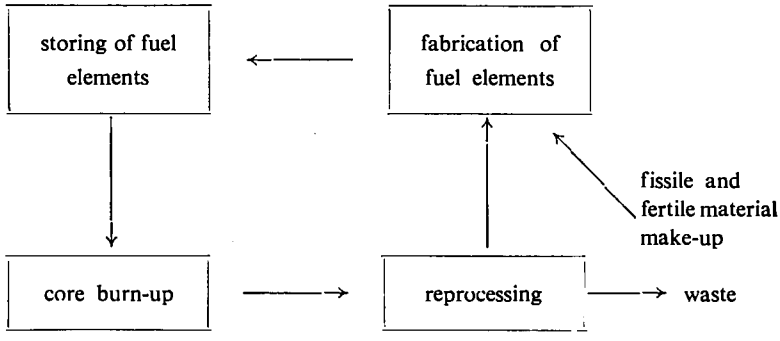
The THTR is a movable fuel type reactor. The fuel elements are balls with dimensions much smaller than the dimensions of the core. The liquid like properties of such a bed of balls allow for recirculating the fuel elements. Criticality at constant core dimensions is achieved by continuous supply of fresh balls and equivalent withdrawing of balls with high burn-up. In this way no excess reactivity has to be built in. Further there are no losses of neutrons in control rods. This procedure presumes the possibility of measuring the burn-up state of the individual fuel elements.

The combined evaluation of burn-up characteristics and fuel cycle costs is done for different initial inventories of fissile and fertile material per ball. The first step in this program is the calculation of the average and the final concentration vectors and of the deduced burn-up parameters like fifa, MWd/t initial fissile and fertile material, conversion ratio etc... The calculations are performed for a steady state model with high recirculation rate of the balls. The last assumption means uniform distribution of all materials in the core. The calculation assumes reprocessing of the heavy materials, whereby U^{233} and U^{235} are reintroduced into the fresh fuel. A 93 % U^{235}-enrichment of the make-up fuel is chosen throughout. In all main cases the initial inventory per ball of U^{235} is held constant (1 g resp. 1.5 g per ball). The varying parameter in all subcases is the ratio of the initial thorium inventory per ball to the U^{235} make-up necessary to get the fixed initial inventory of U^{235} per ball.

The second step in the calculations process is represented by the evaluation of the fuel cycle costs. Several assumptions are made. Investment costs are not

(1) Work done by H. EMMELMANN, W. GERKE, W. GIESSER and H. HUBER.

considered. The costs for (1) the first core load, (2) the first load of the fuel element store, (3) the reprocessing of the final core load and the first store load are assumed to be investment costs. No crediting for the final core fuel is accounted for. The fuel cycle costs are estimated according to the following scheme :



The individual cost positions may be summarized as (1) current costs, (2) interest and taxes for the initial load of a fuel element store, (3) interest and taxes for the initial load of the processing plant. Position (3) may be estimated pessimistically by assuming an equivalenty increased time of delivery of the fresh fuel elements. This procedure is justified by the fact that the main contribution to the fuel cycle costs is given by position (1). The remaining cost positions (1) and (2) show the following fine structure : current costs respectively interest and taxes etc. for (*a*) the fissile material, (*b*) the fertile material, (*c*) the fabrication of fuel elements, (*d*) the reprocessing of the fuel elements with high burn-up.

The results are indicating that the processing costs are of secondary importance. The principal cost positions are the current costs for fuel make-up and fabrication of the fuel elements. Different optimization aspects and restricting conditions like an upper limit of power peaking etc. are to be considered.

2. — Evaluation of Burn-up Characteristics

There are several possibilities of distributing the fissile and fertile material. From the neutron economy point of view for example the separate insertion of fissile and fertile materials into different elements seems to be optimum. The fertile material elements may thus be exposed with a residence time different from that of the fissile material elements. The recirculation of fertile material elements with intervening storage periods for the decay of Pa^{233} would guarantee a good use of fissile material. But there are possibly strong restrictions by the power peaking conditions. Therefore, in this context only the variant with fissile and fertile material inserted into one and the same element is considered.

The fuel element consists of fissile and fertile material and graphite as moderating material. The fission products are assumed not to leave the fuel element.

The calculations are performed for the "equilibrium state", which is defined by the assumption that all core characteristics are independent of time. By the further assumption of a very high recirculation rate it is guaranteed that no spatial variations of core materials do occur. Criticality at constant core dimensions is maintained by continuous supply of fresh fuel elements and equivalent withdrawing of elements with high burn-up. It is easily seen that in this model the time average of the concentration vector in a ball equals the sample average.

This concept usually leads to some difficulties if one considers reprocessing. In this case an equilibrium in the above sense cannot be achieved owing to the non-saturating build-up of U^{236}. These difficulties are avoided in the following fuel management : The pellet of the fuel elements consists of a mixture of graphite and coated particles. Two resp. three types of coated particles are assumed. The fresh particle of type 1 contains only a Th^{232} inventory. The final inventory of the first type together with the 93 % U^{235} fuel make-up is inserted into the type 2 coated particles. Eventually the initial inventory of a type 3 coated particle is formed by the final inventory of the type 2 one. The type 2 resp. 3 coated particles are thrown away. Thus no difficulties in connection with U^{256} occur.

The set of equations which describes the burn-up behaviour of the reactor is represented in Fig. 1. There are in the first place the specific burn-up equations describing the time behaviour of the concentration vector of the materials in a ball.

FIG. 1*a*. — Burn-Up Equations.

$$\dot{N}_{25} = -a_{25}N_{25}$$
$$\dot{N}_{26} = -a_{26}N_{26} + a^c_{25}N_{25}$$
$$\dot{N}_{28} = -a_{28}N_{28}$$
$$\dot{N}_{39} = -(a_{39} + \lambda_{39})N_{39} + a_{28}N_{28}$$
$$\dot{N}_{49} = -a_{49}N_{49} + \lambda_{39}N_{39}$$
$$\dot{N}_{40} = -a_{40}N_{40} + a^c_{49}N_{49}$$
$$\dot{N}_{41} = -(a_{41} + \lambda_{41})N_{41} + a_{40}N_{40}$$
$$\dot{N}_{42} = -a_{42}N_{42} + a^c_{41}N_{41}$$
$$\dot{N}_{02} = -a_{02}N_{02}$$
$$\dot{N}_{13} = -(a_{13} + \lambda_{13})N_{13} + a_{02}N_{02}$$
$$\dot{N}_{23} = -a_{23}N_{23} + \lambda_{13}N_{13}$$
$$\dot{N}_{24} = -a_{24}N_{24} + a^c_{23}N_{23} + a_{13}N_{13}$$

18 additional equations for fission products

Nomenclature.

$a = \bar{\sigma}\varnothing + RS$
$\bar{\sigma}$ = microscopic thermal cross section
R = resonance reaction probability
$\varnothing$ = thermal flux
S = source strength of fission neutrons
λ = decay constant
c = capture
N = number density in a ball

FIG. 1*b*. — Burn-Up Equations.

1. — *Criticality Condition.*

$$S = \varepsilon \sum_j \nu_{fj} a_j^f \bar{N}_j$$

2. — *Neutron Balance Condition.*

$$(p - A)S = \sum_K \bar{N}_K \bar{\sigma}_K \varnothing$$

3. — *Power Normalization.*

$$Q = \sum_j \gamma_j a_j^f \bar{N}_j V_c$$

$$\bar{N} = \frac{1}{t_o} \int_o^{t_o} dt N(t)$$

Nomenclature :

$\bar{N}$ = average concentration in the core
ε = fast fission factor
ν_f = average number of neutrons per fission
γ = mean energy release per fission
p = resonance escape probability
A = leakage
V_c = volume of the reactor core
Q = thermal power
j = index of fissile materials
t_o = residence time of the balls in the reactor core

The solution of these equations yields the final concentration vector in a ball and the mean core concentration vector by the above given procedure. There are two groups of equations. The first group is formed by the heavy material equations, the second one by the fission product equations (18 equations). Neutron absorption and β-decay processes are allowed for.

The next equation is given by the criticality condition which expresses the requirement that each source neutron has to produce just one new source neutron.

A further equation is represented by the neutron balance condition for the mean concentration core. The sources have to equal the sinks. Finally the last equation introduces the power normalisation. The total leakage may be prescribed or calculated.

The whole system of equations is solved by multiple iteration. Some characteristic burn-up quantities are calculated. The corresponding definitions are represented in Fig. 2.

3. — EVALUATION OF THE FUEL CYCLE COSTS

The fuel cycle costs considered here show the following composition :

A. costs for fissile material.
B. costs for fertile material.
C. costs for fabrications of fuel elements.
D. costs for fuel element reprocessing.

FIG. 2. — Deduced Burn-Up Quantities.

$$\text{fifa} = \sum_j \frac{a_j^f \bar{N}_j t_o}{N_{25}(0)}$$

$$\text{fifa}' = \sum_j \frac{a_j^f \bar{N}_j t_o}{N_{25}(0) - N_{25}(t_o)}$$

$$A_2 = \frac{1}{8{,}64 \cdot 10^4} \frac{\sum_j \gamma_j a_j^f \bar{N}_j t_o V_c}{\sum_j M_j(0) + \sum_\mu M\mu(0)} \text{ [MW}d/t\text{]}$$

$$CR = \frac{\lambda_{39}\bar{N}_{39} + a_{40}\bar{N}_{40} + \lambda_{13}\bar{N}_{13}}{\sum_j a_j \bar{N}_j + \lambda_{41}\bar{N}_{41}}$$

Nomenclature :

j = index of fissile materials
μ = index of fertile materials
t_o = residence time of the balls in the reactor core
M = total mass in the reactor core
V_c = volume of the reactor core

All these items subdivide into two positions :

1. current costs.
2. interest and taxes for the first load of the fuel element store.

The corresponding calculation formulas are summarized in Fig. 3. The individual quantities are defined as follows :

E	= net electrical energy output per unit volume of reactor core per unit time	
η	= thermal efficiency	(—)
λ	= load factor	(—)
$\mathscr{L}$	= power density	(W/cm^3)
a, b	= specific costs for fissile resp. fertile material	(DM/g)
e, c	= specific costs for fabrication resp. reprocessing processes	(DM/g heavy material)
t_o	= residence time of the fuel elements in the reactor core	(y)
$N(o)$, $N(t_o)$, $\bar{N}$	= initial, final and mean atomic densities per ball	(cm^{-3})
j	= index of fissile materials	
μ	= index of fertile materials	
S	= index of heavy materials	
A	= mass number	
L	= Loschmidt number	
τ	= time of delivery of fuel elements	(y)
α	= factor to take into account delivery delays	(—)
p	= interest rate	(y^{-1})
s	= tax rate	(y^{-1})

FIG. 3*a*. — Calculation of Fuel Cycle Costs.

A. — FISSILE MATERIALS

1. — *Current Costs*

$$A_1 = \frac{a_{25}}{E}\frac{[N_{25}(0) - N_{25}(t_o)]A_{25}}{L}\frac{1}{t_o}\lambda 10^2$$

2. — *Interest and taxes for the initial load of a fuel element store.*

$$A_2 = \frac{1}{E}\sum_j a_j \frac{N_j(0)A_j}{L}\frac{1}{t_o}\tau(1+\alpha)(p+s)$$

B. — FERTILE MATERIAL

1. — *Current Costs*

$$B_1 = \frac{1}{E}\sum_\mu b_\mu \frac{(N_\mu(0) - N_\mu(t_o))A_\mu}{L}\frac{1}{t_o}\lambda 10^2$$

2. — *Interest and taxes for the initial load of a fuel element store.*

$$B_2 = \frac{1}{E}\sum_\mu b_\mu \frac{N_\mu(0)A_\mu}{L}\frac{1}{t_o}\tau(1+\alpha)(p+s)$$

A_i, B_i [P*f*/kWh]

j, μ = index of fissile resp. fertile materials

For definitions see pages 392-393.

FIG. 3*b*. — Calculation of Fuel Cycle Costs.

C. FABRICATION

1. — *Current Costs*

$$C_1 = \frac{e}{E}\sum_S \frac{N_S(0)A_S}{L}\frac{1}{t_o}\lambda 10^2$$

2. — *Interest and taxes for the initial load of a fuel element store.*

$$C_2 = \frac{e}{E}\sum_S \frac{N_S(0)A_S}{L}\frac{1}{t_o}\tau(1+\alpha)(p+s)$$

D. — REPROCESSING

1. — *Current Costs*

$$D_1 = \frac{c}{E}\sum_S \frac{N_S(t_o)A_S}{L}\frac{1}{t_o}\lambda 10^2$$

2. — *Interest and taxes for the initial load of a fuel element store.*

$$D_2 = \frac{c}{E}\sum_S \frac{N_S(t_o)A_S}{L}\frac{1}{t_o}\tau(1+\alpha)(p+s)$$

$E = 8.762\,\eta\lambda\mathscr{L}$[kWh/cm^3 reactor core]

For Definitions see pages 392-393.

4. — Results

The calculations have been performed with the following set of input data :

Table 1

Notation	Symbol	Dim.	Value
Price per g fissile material	a_{25}	DM/g	48.12
Price per g fertile material	b_{02}	DM/g	0.10
Specific fabrication costs for fuel elements per g heavy material	e	DM/g	4.00 2.00 1.00
Specific reprocessing costs per g	c	DM/g	0.50 0.10
Heavy material			
Interest rate	p	y^{-1}	6.5 %
Tax rate	s	y^{-1}	3.1 %
Load factor	λ	—	0.8
Thermal efficiency	η	—	0.42
Power density	$\mathscr{L}$	W/cm³	7
Time of delivery for the fuel elements	τ	y	0.5
Safety factor for delivery delay	α	—	0.05
Reprocessing time	$\tau_{repr.}$	y	0.5

The results of the calculations are reproduced in Fig. 4 and 5. Fig. 4 and 5 are representing the main cases 1 g resp. 1.5 g U^{235} insert per fresh fuel element. As readily mentioned the considered subcases are characterized by different fertile material (thorium) inventories in a fresh ball. Therefore in the diagrams the ratio of fresh thorium inventory per ball to U^{235} fuel make-up is chosen as abscissa. In the upper half of the diagrams the characteristic burn-up data are represented. The corresponding fuel element costs have been reproduced in the lower half.

There are several possibilities of optimizing reactor fuel cycles. In this report a minimum fuel cycle cost point of view is chosen. It must be mentioned however that in technically feasible cases there are restrictions by the power peaking. Only power peakings $<$ 2-3 are admissible. Thus only a relative optimum can be found.

The cost calculations are performed for different specific fabrication and reprocessing costs. The influence of the last item is seen to be of secondary importance. The main cost contributions are the current costs for fuel make up and the current fabrication costs for the fuel elements. By inspection of the diagrams it can be seen that for high fabrication costs the burn-up parameters fifa $\sim t_0$ resp. fifa′ are no longer characteristic for the total fuel cycle costs.

Finally it may be recognized that for decreasing fabrication costs the total cost minimum is shifted towards higher ThO_2/U_{25}′ ratios.

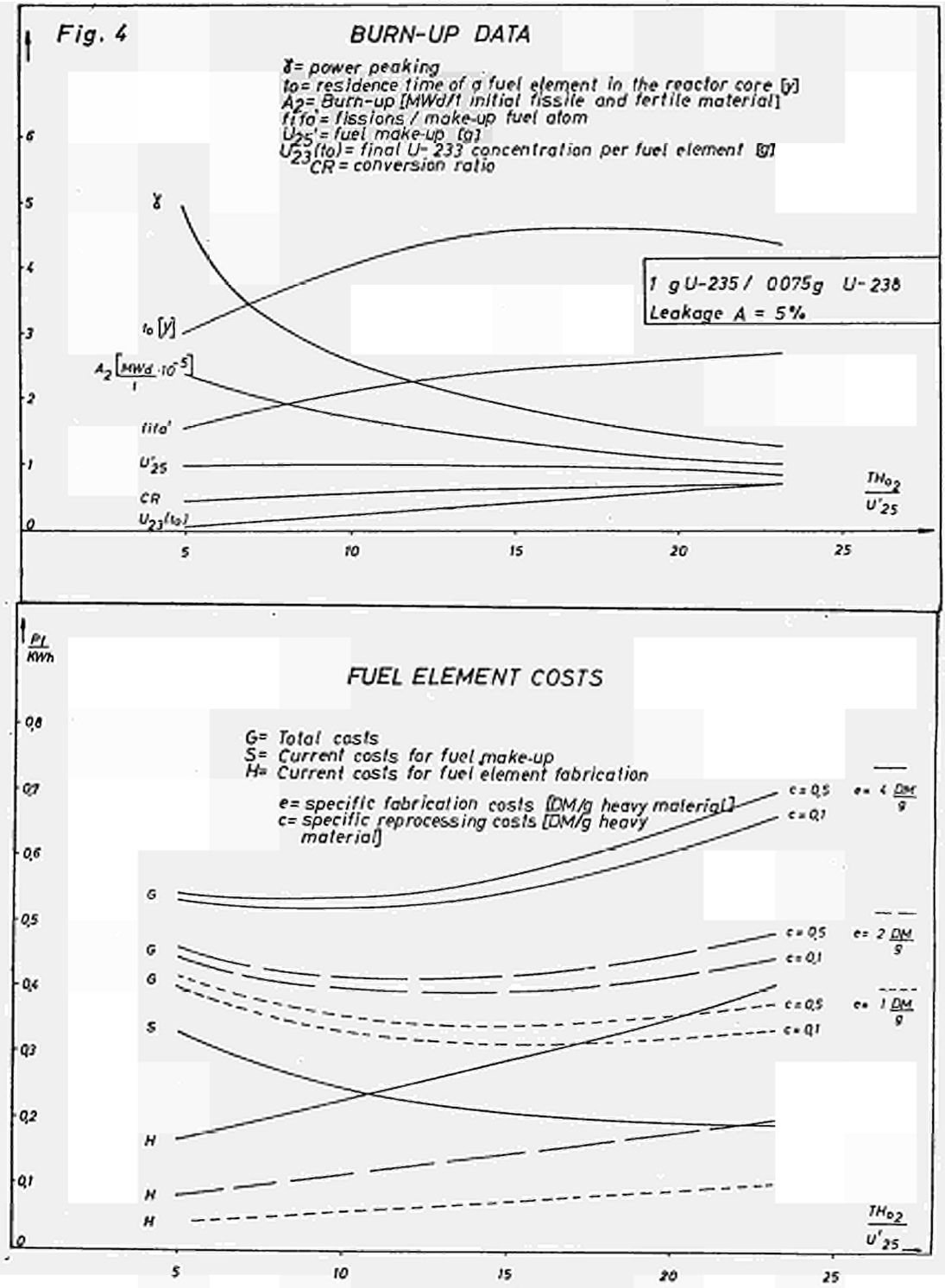


FIG. 4. — Burn-Up Data.

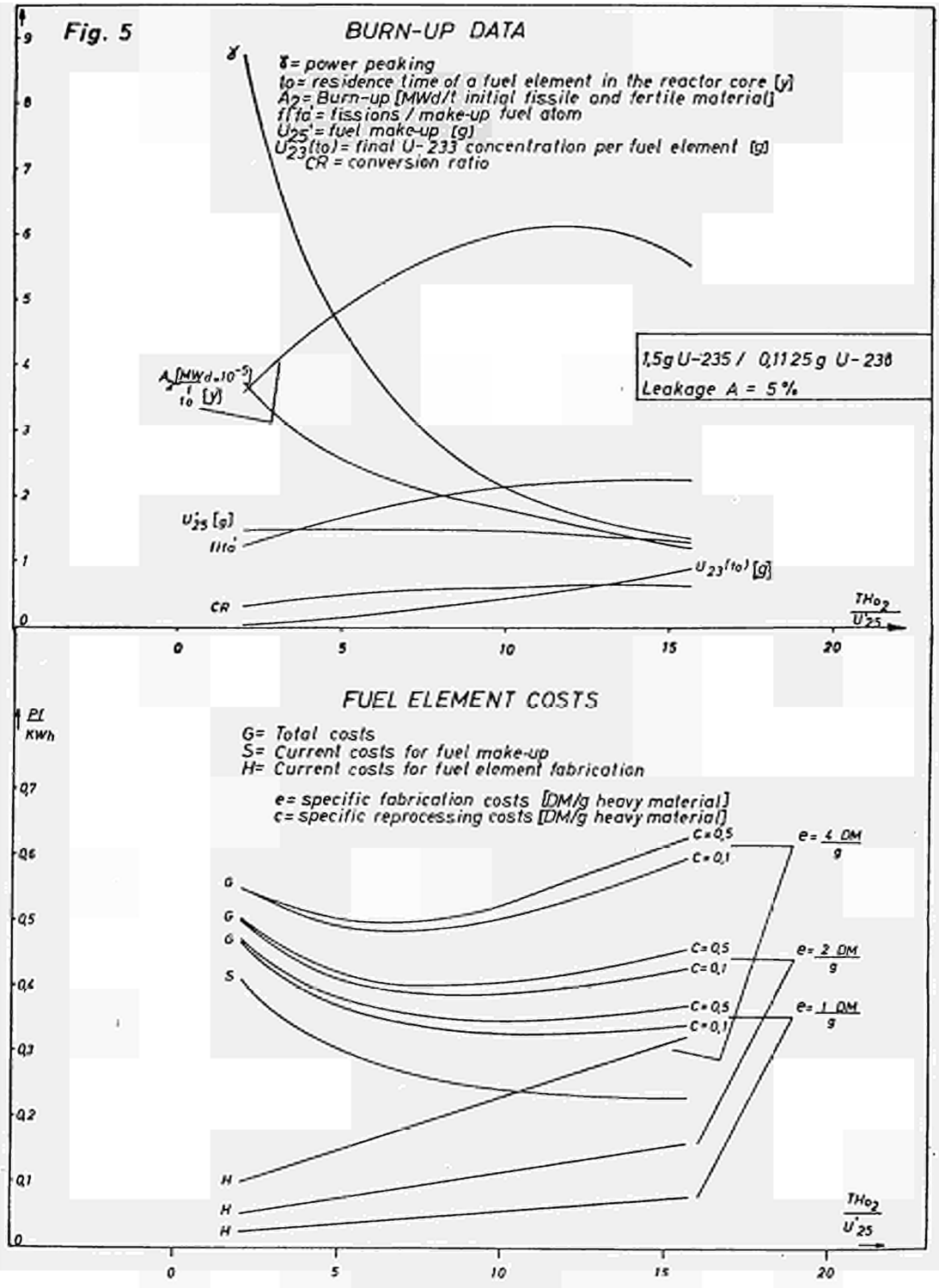


FIG. 5. — Burn-Up Data.

BREEDING IN PEBBLE-BED REACTORS

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ABSTRACT

The conversion factor of a High Temperature Thorium Reactor of the Pebble-Bed Type is given with respect to the produced daughter elements, the poisoning by the fission products and the neutron leakage. The separate consideration of fissile and fertile materials allows the computation of the conversion factor as a function of the distinct life-time of these two element types. For a fuel element life-time of half a year and a breeding element life-time of one year, the conversion factor was calculated as C = 1.04. The proper doubling time for the fissile material is less than 20 years.

The conversion factor of the reactor depends on the concentration of the fission products and hence on the processing frequency as well as on the specific power, the neutron spectrum and the neutron leakage. In the neutron balance the leakage diminishes the number of generated fast neutrons as well as the number of acting slow neutrons; on the other hand, as the cross sections change with the neutron spectrum, the constants taken as a basis for the calculation change, too. The specific power affects the protactinium absorption. The dependence of the conversion factor on the concentration of the fission products is given primarily by the irradiation dose of the fissile and fertile materials.

The spherical fuel and breeder elements pass frequently enough through the reactor during their life-time, so that we may base our calculations upon the concept of flowing fuel and fertile materials. Thus, if the time for a run is short compared to the life-time of the elements, each element is subjected to an average value of all the concentrations involved. Thus, all the concentrations are put into the calculations as values averaged over time. In this way of thinking the mean value by volume is practically replaced by the mean value by time over the whole life-time of the elements. Considering a certain statistical fluctuation, the principle of flowing fuel has the advantage that all the elements receive the same neutron dose. So, the elements attain a regular utilization, and the computation is also simplified.

For our further considerations we introduce the rate s for absorption :

$$s = \int_{U_0}^{U_1} \Phi(U) \cdot \sigma_a(U) \cdot dU \qquad (1)$$

This rate is described as the integrated product of the microscopic absorption cross-section and the neutron flux. The integration is carried over the whole neutron spectrum.

In an appropriate way the pebble-bed reactor containing fertile material is charged with two element types, the fuel and the breeder elements, the fission products generated in the inserted and bred fissile material being built up in different ways. When the reactor is charged, the fuel elements contain merely fissile material, the breeder elements contain thorium as fertile material which generates the daughter elements U^{233} and U^{235} in a saturated concentration. Due to their continuous passing through the reactor, and by the fact of not being separated in the reprocessing, isotopes are present in an equilibrium concentration, too. All the other daughter products, e.g. neptunium, and all the fission products are separated during the reprocessing and hence don't arrive at the equilibrium concentration; so their build-up and their concentrations averaged over their lifetime depend on the dose. The uranium isotopes issued from the reprocessing are used for the fabrication of new fuel elements.

The advantage of the described two-type-loading is that each the fuel and the breeder elements can remain during different times in the reactor. Thus, the total absorption by fission products in the elements is lowered considerably.

In the following description the symbols from Table 1 and 2 are used. All concentrations which are not in the equilibrium state are affected with an upper index number (1) for fuel element, (2) for breeder element.

We start with the neutron balance which is derived from the well known four factor formula. Instead of introducing the absorption in the thermal region only, as it is usually done, we consider the reaction rates over the whole neutron spectrum.

TABLE 1

	Isotope	Rate	Concentration	
	U^{233}	s_3	X_3	
	U^{235}	s_5	X_5	
	Th^{232}	s_2	X_2	
	U^{234}	s_4	X_4	
	U^{236}	s_6	X_6	
	Np^{237}	s_7	X_7	
	Fission products	s_i	X_i	
	Xe^{135}	s_{Xe}	X_{Xe}	
	Pa^{233}	s_{Pa}	X_{Pa}	
	Moderator	s_M	X_M	
	Daughter elements of higher order	s	X_I	

TABLE 2

Γ_s	fast leakage
γ_i	fission product yield
$\eta_{3/5}$	thermal fission neutron yield
Σ_i	sum over all the fission products
B^2	geometric buckling
L_M^2	diffusion length in graphite
$\alpha_{3/5}$	capture-to-fission ratio
$\tau^{1/2}$	lifetime of element (1) respectively (2)
λ	decay constant
q	slowing down density
$\xi\Sigma_s$	slowing down force of the medium
ξ	mean logarithmic energy loss
Σ_s	macroscopic scattering cross section
p	resonance escape probability
Σ_a	macroscopic absorption cross section
Λ/s_3	breeding gain per reaction
RI	resonance integral

With that we write all the terms which belong to the generation of new fuel on the left-hand side of Eq. (2), the other terms being written on the right-hand side.

$$s_2\bar{X}_2 + s_4\bar{X}_4 = \Gamma_s\eta_3 s_3\bar{X}_3 + \Gamma_s\eta_5 s_5\bar{X}_5 - s_3\bar{X}_3 - s_5\bar{X}_5 - s_{Pa}\bar{X}_{Pa} - s_6\bar{X}_6 - \\ - s_7\bar{X}_7^{(1)} - s_7\bar{X}_7^{(2)} - \sum_i s_i\bar{X}_{i3}^{(1)} - \sum_i s_i\bar{X}_{i5}^{(1)} - \sum_i s_i\bar{X}_{i3}^{(2)} - \sum_i s_i\bar{X}_{i5}^{(2)} \\ - s_{Xe}\bar{X}_{Xe3} - s_{Xe}\bar{X}_{Xe5} - s_M\bar{X}_M - DB^2\Phi_{th} \qquad (2)$$

After appropriate transformations the concentrations of reacting matter are generated from the thorium supply. Therefore we can write all the fission and daughter product concentrations in terms of the thorium reaction rate. Let's begin with the concentration of the generated protactinium 233.

$$\frac{dX_{Pa}}{dt} = s_2X_2 - (\lambda_{Pa} + s_{Pa})X_{Pa} \qquad (3)$$

From Eq. (3) we derive the reaction rate of protactinium for the equilibrium state

$$s_{Pa}\bar{X}_{Pa} = \frac{s_{Pa}}{s_{Pa} + \lambda_{Pa}} s_2\bar{X}_2 \qquad (4)$$

For the average concentration of the bred U^{233} we obtain

$$\frac{dX_3}{dt} = \lambda_{Pa}X_{Pa} - (s_3 + \Lambda)X_3 \qquad (5)$$

The bred fuel factor is given by Λ; this fuel can be removed from the reactor system. $\frac{\Lambda}{s_3}$ is the breeding gain of U^{233} based on one reaction. The concentration of U^{233} is given by the following equation :

$$\bar{X}_3 = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{1}{s_3 + \Lambda} \cdot s_2\bar{X}_2 \qquad (6$$

The U^{234} is produced firstly by neutron capture in U^{233} and secondly by beta-decay of Pa^{234}; the latter being produced by neutron capture in Pa^{233}. Thus, we get the following differential form for the concentration of U^{234}

$$\frac{dX_4}{dt} = \frac{\alpha_3}{1+\alpha_3} s_3 X_3 + s_{Pa} X_{Pa} - (s_4 + \Lambda) X_4 \tag{7}$$

Introducing Λ in the equation above we have taken into account that U^{234} is removed from the fuel cycle during the reprocessing. The U^{234} concentration is given in Eq. (8).

$$\bar{X}_4 = \frac{1}{s_4 + \Lambda}\left(\frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_3}{s_3 + \Lambda} + \frac{s_{Pa}}{\lambda_{Pa} + s_{Pa}}\right) s_2 \bar{X}_2 \tag{8}$$

In a similar manner we can figure the produced U^{235} in the following equation :

$$\frac{dX_5}{dt} = s_4 X_4 - (s_5 + \Lambda) \cdot X_5 \tag{9}$$

and

$$\bar{X}_5 = \frac{1}{s_5 + \Lambda} \cdot \frac{s_4}{s_4 + \Lambda}\left(\frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_3}{s_3 + \Lambda} + \frac{s_{Pa}}{\lambda_{Pa} + s_{Pa}}\right) s_2 \bar{X}_2 \tag{10}$$

The concentration of U^{236} is represented by

$$\frac{dX_6}{dt} = \frac{\alpha_5}{1+\alpha_5} s_5 X_5 - (s_6 + \Lambda) X_6 \tag{11}$$

and

$$\bar{X}_6 = \frac{1}{s_6 + \Lambda} \cdot \frac{\alpha_5}{1+\alpha_5} \cdot \frac{s_5}{s_5 + \Lambda} \cdot \frac{s_4}{s_4 + \Lambda}\left(\frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_3}{s_3 + \Lambda} + \frac{s_{Pa}}{s_{Pa} + \lambda_{Pa}}\right) s_2 \bar{X}_2 \tag{12}$$

Because of the short half-life of U^{237}, which is generated by neutron capture in U^{236}, we may assume in our calculations that the transition from U^{236} to Np^{237} is a spontaneous one. The neptunium being removed from the fuel cycle during the reprocessing, we must compute the reaction rate of Np separately for the elements (1) and (2).

The change of the Np-concentration is given by

$$\frac{dX_7}{dt} = s_6 X_6 - s_7 X_7 \tag{13}$$

This leads us to the reaction rate for element (1)

$$s_7 \bar{X}_7^{(1)} = \frac{\alpha_5}{1+\alpha_5}\left[1 - \frac{1 - e^{-s_7 \tau^1}}{s_7 \tau^1}\right] s_5 \bar{X}_5^{(1)} \tag{14}$$

and for element (2)

$$s_7 \bar{X}_7^{(2)} = \frac{\alpha_5}{1+\alpha_5}\left[1 - \frac{1 - e^{-s_7 \tau^2}}{s_7 \tau^2}\right] s_5 \bar{X}_5^{(2)} \tag{15}$$

Because of its delayed build-up and its relatively fast elimination during the reprocessing the Np^{238} has no significant influence on the neutron balance and therefore will not be considered anymore in our further calculations.

The concentration of the fission-products depends on the dose, too. As for neptunium we will calculate their reaction rates separately for the two element types, and so we will be able to predict the fission-product poisoning for a different life-time of the two elements in the reactor. Using the poison-functions shown in the Appendix, we obtain the reaction rates of the fission-products issued from U^{233} and U^{235} in the elements (1) and (2) :

$$\sum_t s_t \bar{X}_{t3}^{(1)} = V_{sp}(s_3\tau^1) \cdot s_3\bar{X}_3^{(1)} \tag{16}$$

$$\sum_t s_t \bar{X}_{t5}^{(1)} = V_{sp}(s_5\tau^1) \cdot s_5\bar{X}_5^{(1)} \tag{17}$$

$$\sum_t s_t \bar{X}_{t3}^{(2)} = V_b(s_3\tau^2) \cdot \frac{s_3\bar{X}_3^{(2)}}{1 - \dfrac{1 - e^{-s_3\tau^2}}{s_3\tau^2}} \tag{18}$$

$$\sum_t s_t \bar{X}_{t5}^{(2)} = V_b(s_5\tau^2) \cdot \frac{s_5\bar{X}_5^{(2)}}{1 - \dfrac{1 - e^{-s_3\tau^2}}{s_3\tau^2}} \tag{19}$$

The fuel reaction rates in Eq. (14) to (19) can be written with respect to $s_2\bar{X}_2$ if we use the following equation :

$$\frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} s_2\bar{X}_2 = s_3\bar{X}_3^{(1)} + s_3\bar{X}_3^{(2)} \tag{20}$$

This equation means that the U^{233} produced in the breeder elements is consumed either in the breeder elements themselves or in the fuel elements. The part Λ, i.e. the breeding gain withdrawn from the fuel cycle, is not taken in account in Eq. (20). Despite the accuracy of 5 % we however may use this approach for the transformations of Eq. (16) to (19), the very poison-functions being known within an accuracy of some $\pm$ 15 % due to the rather inaccurate knowledge of the cross-sections and resonance-integrals for the fission-products. So far as the fission-products are concerned, they absorb some 2 to 5 % of all the generated neutrons hence the conversion ratio suffers an error of about 0.5 to 1 % by these inaccuracies. For the build-up of U^{233} in the elements (2) we may write

$$X_3^{(2)} = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{1}{s_3} s_2\bar{X}_2 \,(1 - e^{-s_3 t}) \tag{21}$$

provided the Th-concentration is assumed constant. The corresponding reaction rate, averaged over the life-time of element (2) is given by

$$s_3\bar{X}_3^{(2)} = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} s_2\bar{X}_2 \left(1 - \frac{1 - e^{-s_3\tau^2}}{s_3\tau^2}\right) \tag{22}$$

Using Eq. (20) we get for the element (1)

$$s_3\bar{X}_3^{(1)} = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} s_2\bar{X}_2 \left(\frac{1 - e^{-s_3\tau^2}}{s_3\tau^2}\right) \tag{23}$$

Multiplying Eqs. (22) and (23) by $\frac{\alpha_3}{1+\alpha_3}$ we get the U^{235}-absorption rate in the elements (1) and (2) as a pessimistic approximation

$$s_5\bar{X}_5^{(1)} = \frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} s_2\bar{X}_2 \frac{1 - e^{-s_3\tau^2}}{s_3\tau^2} \tag{24}$$

and

$$s_5\bar{X}_5^{(2)} = \frac{\alpha_3}{1+\alpha_3} s_3\bar{X}_3^{(2)} + \frac{s_{Pa}}{\lambda_{Pa} + s_{Pa}} s_2\bar{X}_2 \tag{25}$$

$$= s_2\bar{X}_2 \left[\frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \left(1 - \frac{1 - e^{-s_3\tau^2}}{s_3\tau^2}\right) + \frac{s_{Pa}}{\lambda_{Pa} + s_{Pa}}\right]$$

The produced U^{235} by n-capture in Pa^{233} is taken into consideration in the second term of Eq. (25). Consequently all the neptunium and fission-product parts are figured as a function of the thorium-reaction rate.

The reaction rates of xenon-poisoning can be immediately deduced from Eqs. (6) and (10) by forming the appropriate equilibrium concentrations.

$$s_{Xe}\bar{X}_{Xe3} + s_{Xe}\bar{X}_{Xe5} = \left(\frac{\gamma_{Xe3}}{1+\alpha_3} s_3\bar{X}_3 + \frac{\gamma_{Xe5}}{1+\alpha_5} s_5\bar{X}_5\right) \frac{s_{Xe}}{s_{Xe} + \lambda_{Xe}}$$

$$= \left[\frac{\gamma_{Xe3}}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_3}{s_3 + \Lambda} + \frac{\gamma_{Xe5}}{1+\alpha_5} \cdot \frac{s_5}{s_5 + \Lambda} \cdot \frac{s_4}{s_4 + \Lambda}\right.$$

$$\left. \cdot \left(\frac{\alpha_3}{1+\alpha_3} \cdot \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_3}{s_3 + \Lambda} + \frac{s_{Pa}}{s_{Pa} + \lambda_{Pa}}\right)\right] \frac{s_{Xe}}{s_{Xe} + \lambda_{Xe}} s_2\bar{X}_2 \tag{26}$$

In the determination of the Pa-losses, Eq. (3), we neglected that a decrease of this fraction becomes possible by passing the fuel elements through the reactor with a period in the order of magnitude of the protactinium half-life. Preliminary calculations have shown that this procedure is able to reduce the fraction mentioned above to somewhat 70-80 %.

Thus the terms of Eq. (2) are all figured as a function of the thorium reaction rate. Dividing both sides of Eq. (2) by the sum of the reaction rates of the two fuel fractions, we obtain the conversion factor. The protactinium-loss during the transformation of fertile material to fissile material is taken into account by multiplying the conversion factor by the coefficient $\frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}}$. The pessimistic error which arises in that we multiply the producing rate of new fuel generated by the conversion of U^{234} to U^{235} by this coefficient, too, is so insignificant, that we may neglect this error in order to simplify the calculations. Thus, the conversion factor can be written in the following form :

$$C = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \cdot \frac{s_2\bar{X}_2 + s_4\bar{X}_4}{s_3\bar{X}_3 + s_5\bar{X}_5} \tag{27}$$

1. — RESULTS

For the numerical evaluation of the preceding formulae we must note that the rate s of the different nuclei always appears as a ratio of two rates, respectively, that each rate can be based on the rate of U^{233}.

$$s_i = \frac{s_i}{s_3} \cdot s_3 \tag{28}$$

The influence of the spectrum is taken into account by computing the rates in the following way :

$$s = (\sigma_{th} + B \cdot RI) \cdot \Phi_{th} \tag{29}$$

The factor B is a characteristic worth for the hardness of the spectrum and has been calculated approximately to 0.20-0.15 for a moderator-to-uranium ratio of 7,000 to 10,000. These values are close to those indicated by Chernick[1]. The error that occurs when writing the rates in the way above affects nuclei with a rather great resonance integral only, e.g. protactinium. In the form Eqs. (28) and (29) is written, the flux is contained only in the rate s_3, the absolute value of which is a parameter in the calculation and must be taken from experimental data.

For calculations of temperature dependence of the rates s, we bring Eq. (1) in the form :

$$s_i = \sigma_{th} \cdot \Phi_{th} + \int_{U_1}^{U_2} \Phi(U)\sigma_i(U) \cdot dU \tag{30}$$

where

$$\Phi(U) \cong \text{const} \cong \frac{q}{\xi\Sigma_s} = \frac{\Sigma_a \cdot \Phi_{th}}{\xi\Sigma_s p\Gamma_s} \tag{31}$$

Hence the rate

$$s_i = \sigma_{ith} \cdot \Phi_{th} + RI_i \frac{\Sigma_a \cdot \Phi_{th}}{\xi\Sigma_s p\Gamma_s} \tag{32}$$

In the presence of a $\frac{1}{E}$ - spectrum and as long as we have a $\frac{1}{v}$ - absorption in the thermal region all the s_i - values are independent of temperature in this approximation. For the calculation of η_{eff} we use experimental data from literature (Table 3). With the thermal values for η_o we obtain the η_{eff}-values for U^{233} and U^{235} with an error of about $\pm$ 0.35 %.

$$\eta_{eff} = \eta_o \left[1 - \frac{B \cdot RI}{\sigma_{th} + B \cdot RI}\left(\frac{\alpha_{epi} - \alpha_{th}}{1 + \alpha_{epi}}\right)\right] \tag{33}$$

The value of $\Gamma_s = 0.99$ for the leakage in the fast region for a big reactor (1,000 MWe) with blanket was assumed.

TABLE 3

	U^{233}	U^{235}
η_o	2.292 ± 0.008[2]	2.074 ± 0.006[2]
α_{th}	0.100[3]	0.188[3]
α_{epi}	0.165[4]	0.485[5]

The results based on the foregoing assumptions and data are listed in Table 4 and plotted in Fig. 1; each term in Eq. (2) has been divided by the sum of the fuel-reaction rates. The terms of the conversion factor of the elements which are in the state of saturation are listed in Table 4. The calculations were carried out for an extraction factor $\Lambda = 0$ and $\Lambda = 0.05 \cdot s_3$, and for a U^{233}-rate of $s_3 = 1\ a^{-1}$ and $1.5\ a^{-1}$. In the lowest line we may find the sum of the constant fractions multiplied by $\dfrac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}}$

$$C' = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \left[\frac{\Gamma_s(\eta_3 s_3 \bar{X}_3 + \eta_5 s_5 \bar{X}_5) - s_3 \bar{X}_3 - s_5 \bar{X}_5 - s_{Pa} \bar{X}_{Pa}}{s_3 \bar{X}_3 + s_5 \bar{X}_5} + \frac{- s_6 \bar{X}_6 - s_{Xe} \bar{X}_{Xe3} - s_{Xe} \bar{X}_{Xe5} - s_M \bar{X}_M - DB^2 \Phi_{th}}{s_3 \bar{X}_3 + s_5 \bar{X}_5} \right] \quad (34)$$

In Fig. (1) the dose-dependent fractions of the fission products and neptunium, V_{tot}, are summarized :

$$V_{tot} = \frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}} \frac{s_7 \bar{X}_7^{(1)} + s_7 \bar{X}_7^{(2)} + \sum_i s_i \bar{X}_{i3}^{(1)} + \sum_i s_i \bar{X}_{i5}^{(1)} + \sum_i s_i \bar{X}_{i3}^{(2)} + \sum_i s_i \bar{X}_{i5}^{(2)}}{s_3 \bar{X}_3 + s_5 \bar{X}_5} \quad (35)$$

V_{tot} is plotted vs. the dose $s_3\tau^2$ of the breeder elements and the dose $s_3\tau^1$ of the fuel elements as a parameter. Additionally we plotted on the abscissa the burn-up of the breeder elements in MWd/t.

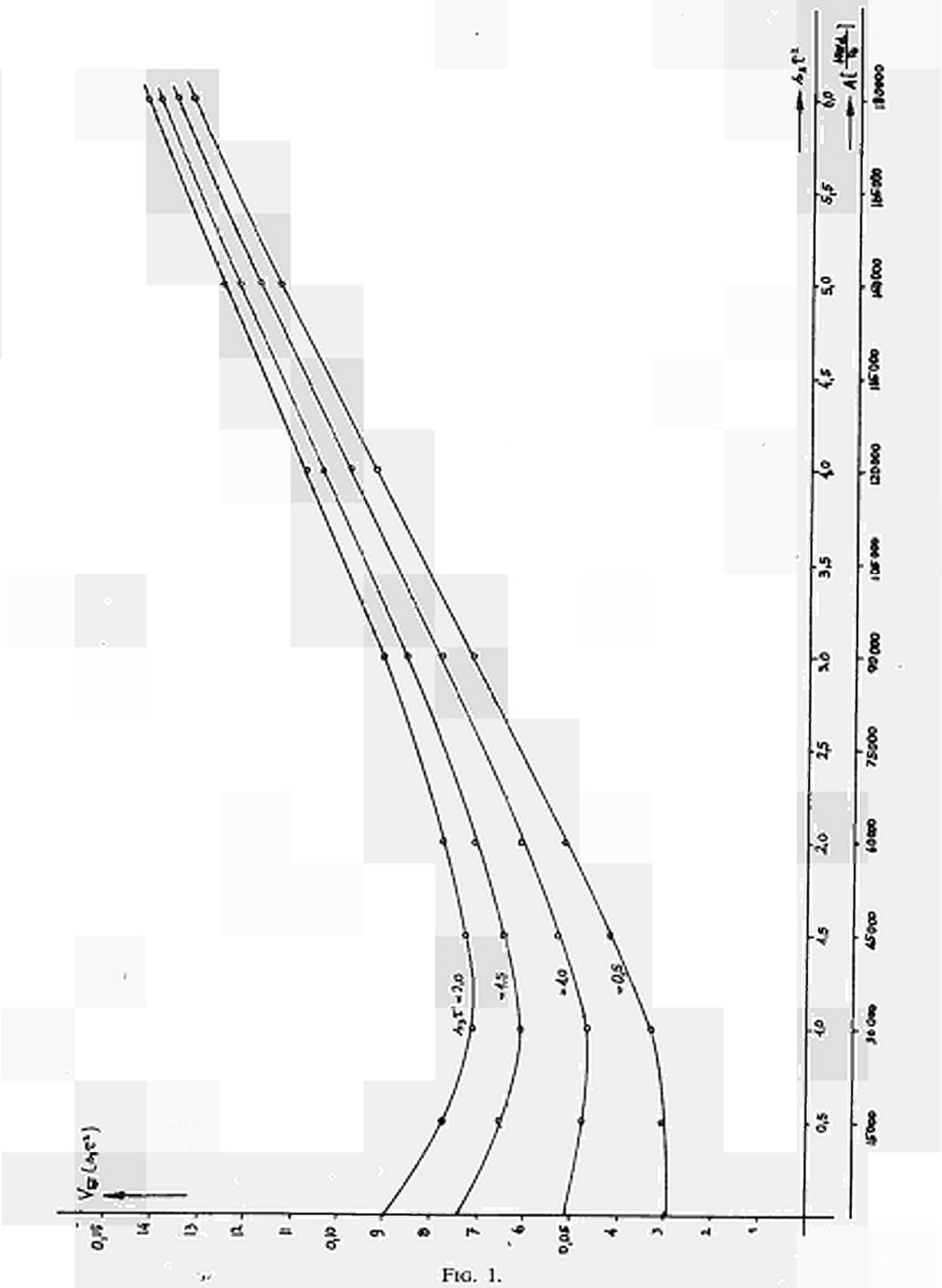

FIG. 1.

TABLE 4

$\Lambda/s_3 = 0.05$

s_3	$1a^{-1}$				$1.5a^{-1}$			
B	0.205	0.182	0.164	0.149	0.205	0.182	0.164	0.149
Γ_{seff}^{-1}	1.213	1.217	1.218	1.224	1.211	1.211	1.217	1.222
—Pa^{233}	0.031	0.029	0.028	0.026	0.039	0.037	0.035	0.033
—U^{236}	0.012	0.012	0.011	0.010	0.013	0.012	0.012	0.011
—Xe^{135}	0.042	0.042	0.042	0.042	0.044	0.044	0.045	0.045
—X_M-leakage . .	0.034	0.041	0.047	0.053	0.034	0.040	0.046	0.053
C′	1.058	1.059	1.060	1.063	1.034	1.036	1.038	1.042

$\Lambda/s_3 = 0$

s_3	$1a^{-1}$				$1.5a^{-1}$			
B	0.205	0.182	0.164	0.149	0.205	0.182	0.164	0.149
Γ_{seff}^{-1}	1.208	1.210	1.213	1.218	1.208	1.208	1.211	1.216
—Pa^{233}	0.029	0.027	0.026	0.024	0.037	0.034	0.033	0.031
—U^{236}	0.021	0.021	0.020	0.020	0.023	0.022	0.021	0.021
—Xe^{135}	0.042	0.042	0.042	0.042	0.044	0.044	0.044	0.045
—X_M-leakage . .	0.034	0.040	0.046	0.052	0.033	0.039	0.046	0.052
C′	1.047	1.048	1.049	1.051	1.028	1.028	1.029	1.032

2. — DISCUSSION

Table 4 shows that in the case of $\Lambda = 0.05 \cdot s_3$ and $s_3 = 1\ a^{-1}$ and with a moderator-ratio of about 10,000 we get for the factor $C' = 1.063$. According to Fig. 1 the total dose-dependent poisoning is 0.033 if the fuel elements have a life-time of half a year and the breeder elements one of one year ($s_3 = 1\ a^{-1}$), viz. a burn-up of the breeder elements of some 30,000 MWd/t. With respect to the foregoing life-time of the elements we obtain a conversion factor of $C = 1.030$. This conversion factor can be enlarged if we consider that the Pa-reaction rate we used for the equi-

librium state concentration was a pessimistic one. In reality, the protactinium is built up only after the reprocessing :

$$X_{Pa} = \frac{s_2 \bar{X}_2}{s_{Pa} + \lambda_{Pa}} \left(1 - e^{-(s_{Pa}+\lambda_{Pa})t}\right) \tag{36}$$

and

$$\bar{X}_{Pa} = \frac{s_2 \bar{X}_2}{s_{Pa} + \lambda_{Pa}} \left(1 - \frac{1}{(s_{Pa} + \lambda_{Pa})\tau} (1 - e^{-(s_{Pa}+\lambda_{Pa})\tau})\right) \tag{37}$$

Hence for an element life-time of one year we get a decrease in the Pa-absorption rate by a factor of 0.89. Due to the Pa-losses when reprocessing we may, in the case of a yearly reprocessing, multiply the Pa-fraction by an extra 0.9 factor. We can reach additional decrease of the Pa-fraction if the elements pass through the reactor in an optimum way; if the passing through-time is in the order of magnitude of the Pa-half-life, and if we take into account the local flux distribution, we obtain still another factor of at least 0.85. Due to these three factors the original Pa-fraction of about 0.052 goes down to 0.035 (in our example the factor $\frac{\lambda_{Pa}}{\lambda_{Pa} + s_{Pa}}$ represents essentially a doubling of the Pa-fraction), viz. the conversion factor goes up to 1.047 by an increase of 0.017. If we suppose the reprocessing losses in an aqueous process to be 0.5 %, we get a final conversion factor of C = 1.04. The proper doubling time is 17 years. With 10 % hold-up in the outer parts of the fuel cycle is this time not more than 20 years.

We would like to mention another possibility to improve the neutron balance : the use of BeO as material for the sphere kernel. A rough estimate showed that considering the additional losses — the Li^6-poisoning could be eliminated by a short-time heating of the elements — the $(n, 2n)$-reaction could improve the conversion factor by at least 0.02.

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5. L. J. Esch *et al.* — *Trans. Am. Nucl. Soc.*, 7 : 78 (1964).
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APPENDIX

In general, the poisoning of a reactor by fission products can be calculated with a good approximation in the following way. The differential equations

$$\frac{dX_i^{(1)}}{dt} = \frac{\gamma_i}{1 + \alpha} s_{sp} X_{sp_0} \cdot e^{-s_{sp}t} - s_i X_i \tag{38}$$

$$\frac{dX_i^{(2)}}{dt} = \frac{\gamma_i}{1+\alpha} s_b X_b (1 - e^{-s_{sp}t}) - s_i X_i \tag{39}$$

are the initial equations for the fission products change in concentration, when the fuel concentration decreases, index (1), Eq. (38), and when the fission products buildup by the breedings, index (2), Eq. (39). X is the concentration of the materials involved; *sp*, *b*, and *i* are the indices for fissile material, fertile material, and for the *i*-th fission product.

The solutions of the foregoing equations are :

$$X_i^{(1)} = \frac{\gamma_i}{1+\alpha} \cdot \frac{1}{s_i - s_{sp}} (e^{-s_{sp}t} - e^{s_i t}) s_{sp} X_{sp_o} \tag{40}$$

$$X_i^{(2)} = \frac{\gamma_i}{1+\alpha} \left[\frac{1 - e^{-s_i t}}{s_i} - \frac{1}{s_i - s_{sp}} (e^{-s_{sp}t} - e^{-s_i t}) \right] s_b X_b \tag{41}$$

If using Eqs. (40) and (41), we average the reaction rates of the fission products $s_i X_i$ over the life time τ of the elements and if we sum all the fission products, we obtain

$$\sum_i s_i \bar{X}_i^{(1)} = s_{sp} \bar{X}_{sp} \sum_i \frac{\gamma_i}{1+\alpha} \frac{s_{sp}}{s_{sp} - s_i} \left(\frac{1 - e^{-s_i \tau}}{1 - e^{-s_{sp}\tau}} - \frac{s_i}{s_{sp}} \right) \tag{42}$$

$$\sum_i s_i \bar{X}_i^{(2)} = s_b \bar{X}_b \sum_i \frac{\gamma_i}{1+\alpha} \left[1 - \frac{s_i}{s_{sp}} \left(1 - \frac{1 - e^{-s_{sp}\tau}}{s_{sp}\tau} \right) - \frac{1 - e^{-s_i \tau}}{s_i \tau} \right] \frac{s_{sp}}{s_{sp} - s_i} \tag{43}$$

In Eq. (43) is, analogous to Eq. (22),

$$s_b \bar{X}_b = \frac{s_{sp} \bar{X}_{sp}}{1 - \dfrac{1 - e^{-s_{sp}\tau}}{s_{sp}\tau}} \tag{44}$$

The abbreviation $r_i = s_i / s_{sp}$ allows us to represent the sums as a function of the element dose τs_{sp}. Thus, the sums are ratios of the fission products absorption to the fissile respectively fertile materials as a function of the elements dose. We shall call them the "Poisoning functions". For a decreasing fuel concentration and an increasing one by breeding the poisoning functions are

$$V_{sp}(s_{sp}\tau) = \sum_i \frac{\gamma_i}{1+\alpha} \frac{1}{1 - r_i} \left(\frac{1 - e^{-r_i s_{sp}\tau}}{1 - e^{-s_{sp}\tau}} - r_i \right) \tag{45}$$

$$V_b(s_{sp}\tau) = \sum_i \frac{\gamma_i}{1+\alpha} \frac{1}{1 - r_i} \left[1 - \frac{1 - e^{-r_i s_{sp}\tau}}{r_i s_{sp}\tau} - r_i \left(1 - \frac{1 - e^{-s_{sp}\tau}}{s_{sp}\tau} \right) \right] \tag{46}$$

These functions have been calculated under the same assumptions as in the principal part of this paper. The factor B for the rate was assumed 0.1 and 0.2. The fission yields γ_i and the cross sections for the fission products are quoted from the paper of Garrison and Roos [6]. Because of the inexact knowledge of these cross sections the accuracy of the poisoning functions is merely some $\pm$ 15 %. The poisoning functions $V_{sp}(s\tau)$ and $V_b(s\tau)$ for U^{233} and U^{235} are plotted in Figs. 2 and 3.

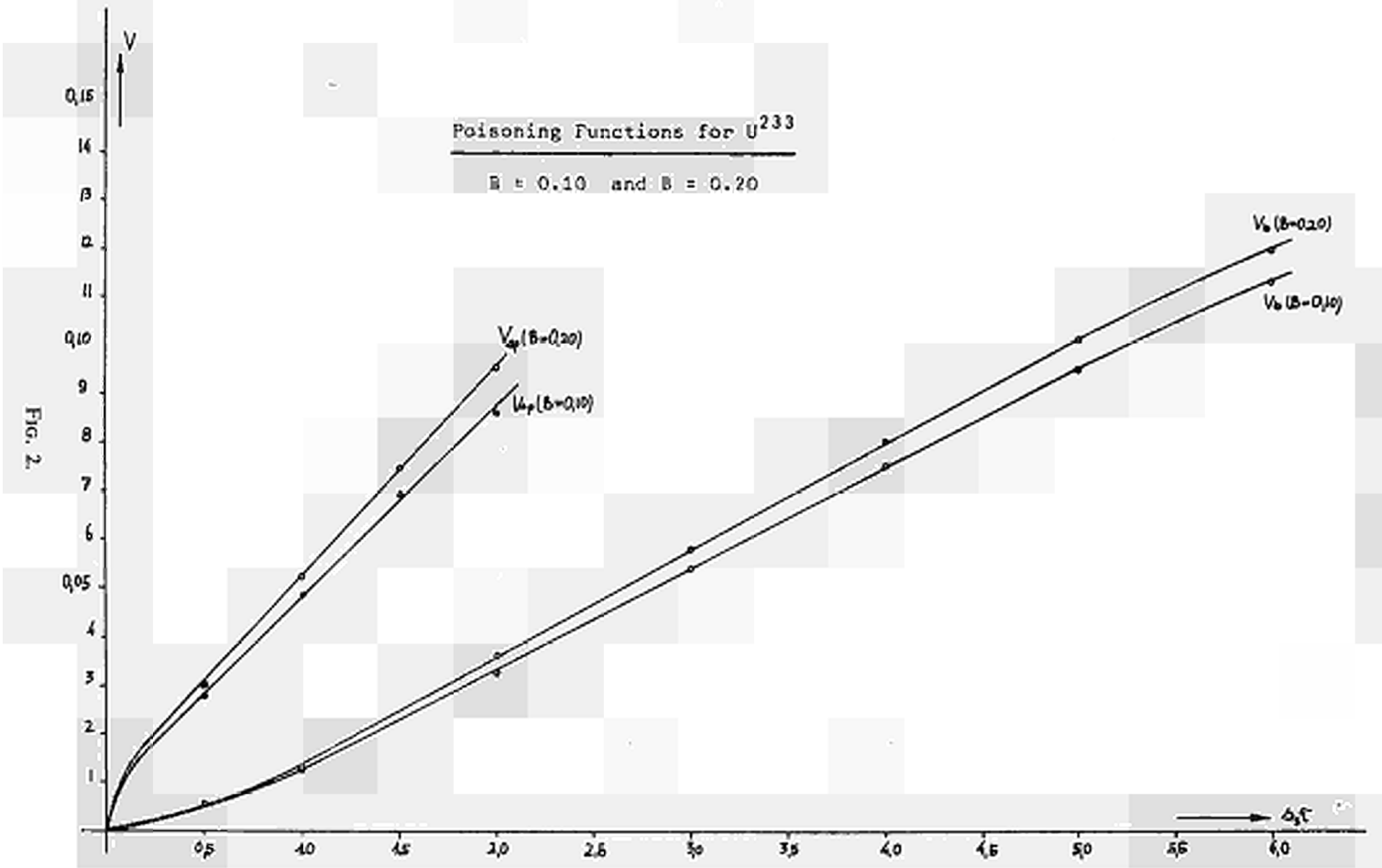
Poisoning Functions for U233
B = 0.10 and B = 0.20
V
0,15
14
13
12
11
0,10
9
8
7
6
0,05
4
3
2
1
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0
5,5
6,0
Vp (B=0,20)
Vp (B=0,10)
Vb (B=0,20)
Vb (B=0,10)

Fig. 2.

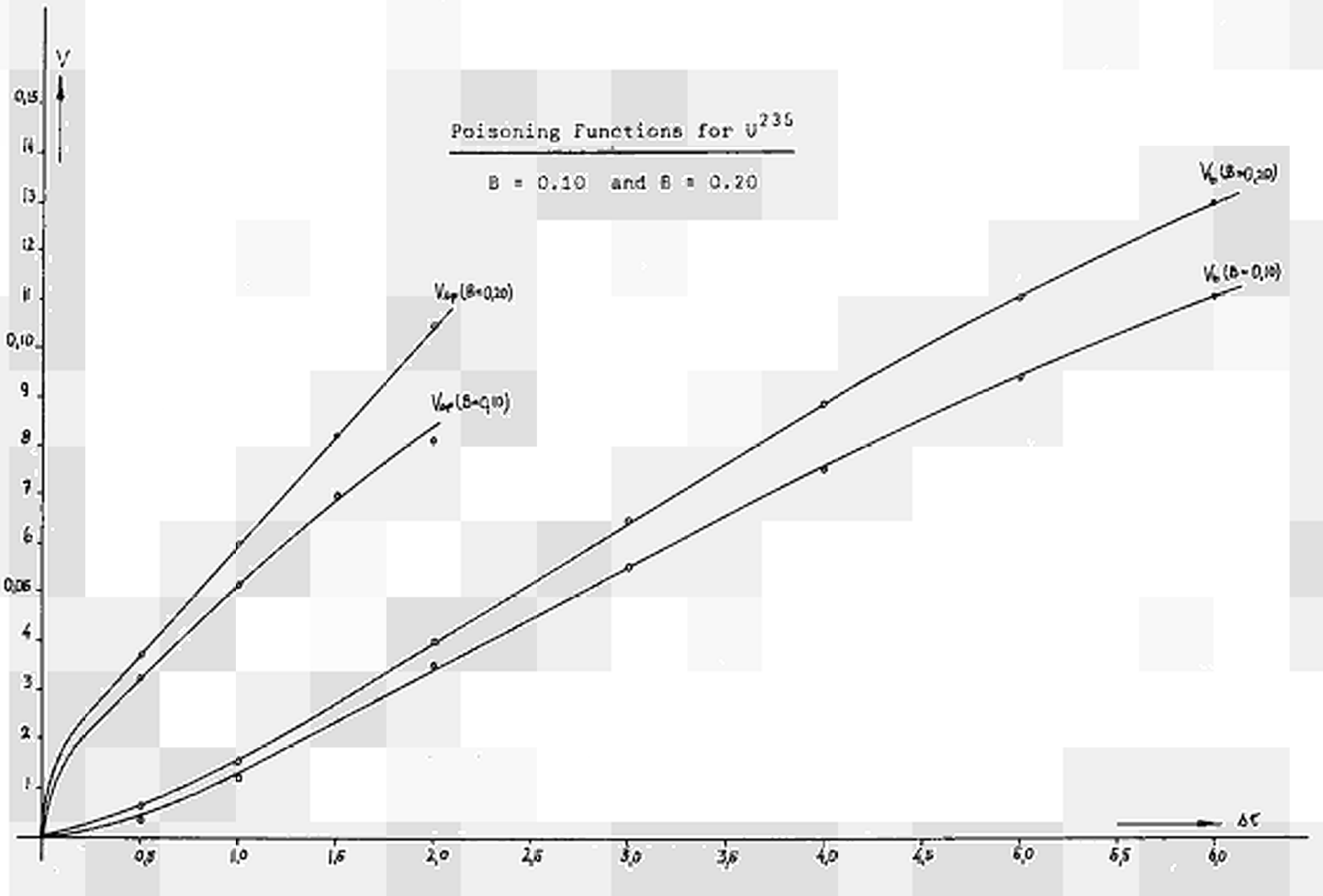
Poisoning Functions for U235
B = 0.10 and B = 0.20
V
0,15
14
13
12
11
0,10
9
8
7
6
0,05
4
3
2
1
V_{eff} (B=0,20)
V_{eff} (B=0,10)
V_0 (B=0,20)
V_0 (B=0,10)
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0
5,5
6,0
δτ

FIG. 3.

FUTURE FUEL REQUIREMENTS FOR ATOMIC POWER PLANTS

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ABSTRACT

This study will give relations between the available reserves of fissile material and the expected requirements of nuclear energy. The fuel amount necessary to cover these energy requirements will be studied without taking into consideration certain reactor types. Calculational parameters will be : specific power, conversion ratio or doubling time, date at which breeders will be available. The results are given as diagrams.

1. — INTRODUCTION

As known, world energy requirements are steadily increasing. Due to limited reserves of conventional energy, it seems a necessity to cover in future a steadily increasing part of the whole energy requirements by nuclear energy. For economic reasons, efforts should be undertaken to handle low-cost fissile material as long as possible. However, since there exists a limitation of these reserves, it seems necessary to change over to economical breeder systems at a certain date. This study should determine relations between energy requirements and uranium existence. The investigation cannot give statements concerning the economy of certain reactor types and the optimal distribution of certain reactor types, to cover nuclear power demand; there is no exact information available, e.g. costs and time of reprocessing, which influence the economy of all reactors (with recycle of bred fuel) to a large extent, to determine such prognosis without considerable mistakes. This influence can be different from the structure of costs of the reactor in question.

To avoid these difficulties — at least in a first stage — special considerations of differential reactors were neglected due to a paper of Kerlin. The reactors are merely characterized by both specific power and conversion ratio or doubling time. Another important size is the date at which breeders will be available.

2. — COMBINATION OF REQUIREMENTS

Starting from the view point that converters are constructed in the first state and after a certain period of time there will be a change-over to breeder reactors, the total fuel amount required consists of four constituents :

1. fuel by which converters are equipped before change-over to breeders;
2. fuel to replace the burned fuel in converters before change-over to breeders;

3. fuel by which breeders are equipped after change-over from converters;
4. fuel to replace the burned fuel in converters after change-over to breeders.

For constituent 4., a converter lifetime of 30 years was estimated.

3. — Fuel Reserves

Various authors estimated the available reserves of fissile material. The results of Kaufmann and Jordan, McKelvey, Lane, Faulkner, and McVey are given in Table I. The amounts of 10^9 short tons U_3O_8 of available reserves, the mining costs of which run up to 500$ per *lb* U_3O_8, were neglected since they are of no importance for the investigations given here. For mining reserves at less than 10$ per *lb* U_3O_8, values given by Kaufmann and Jordan, Lane and Faulkner, and McVey increase from 520,000 to 660,000 short tons U_3O_8; McKelvey's estimations for this category increase by factor 5. Future development will show whether such a prognosis can be justified.

The estimations of Faulkner and McVey are in accordance with the latest results and therefore applied as basis for the following considerations.

Table I. — Estimated Uranium Reserves as a Function of Cost

Cost $ / lbU_3O_8	Resources	Kaufmann/ Jordan 1960		McKelvey 1961		Lane 1962		Faulkner/ McVey 1964	
		Short tons U_3O_8 *1,000	kg U_5 *10^6	Short tons U_3O_8 *1,000	kg U_5 *10^6	Short tons U_3O_8 *1,000	kg U_5 *10^6	Short tons U_3O_8 *1,000	kg U_5 *10^6
<10	Reasonably assured	230	1.27	660	3.64			270	1.49
	Possible additional	300	1.66	1,870	10.32			250	1.38
	Total	530	2.93	2,530	13.96	660	3.64	520	2.87
10-30	Reasonably assured							400	2.21
	Possible additional							300	1.66
	Total					770	4.25	700	3.87
30-50	Reasonably assured	6,000	33.0					5,000	27.61
	Possible additional	6,000	33.0					3,000	16.57
	Total	12,000	66.0	7,700	42.52	3,300	18.22	8,000	44.18
50-100	Reasonably assured							6,000	33.0
	Possible additional							9,000	49.7
	Total							15,000	82.7

4. — Energy and Fuel Requirement

A considerable amount of steadily increasing energy requirements is to be covered by nuclear energy in the future. Different authors have made prognosi of the future demand of nuclear power for the U.S.A.; however, these prognosi show

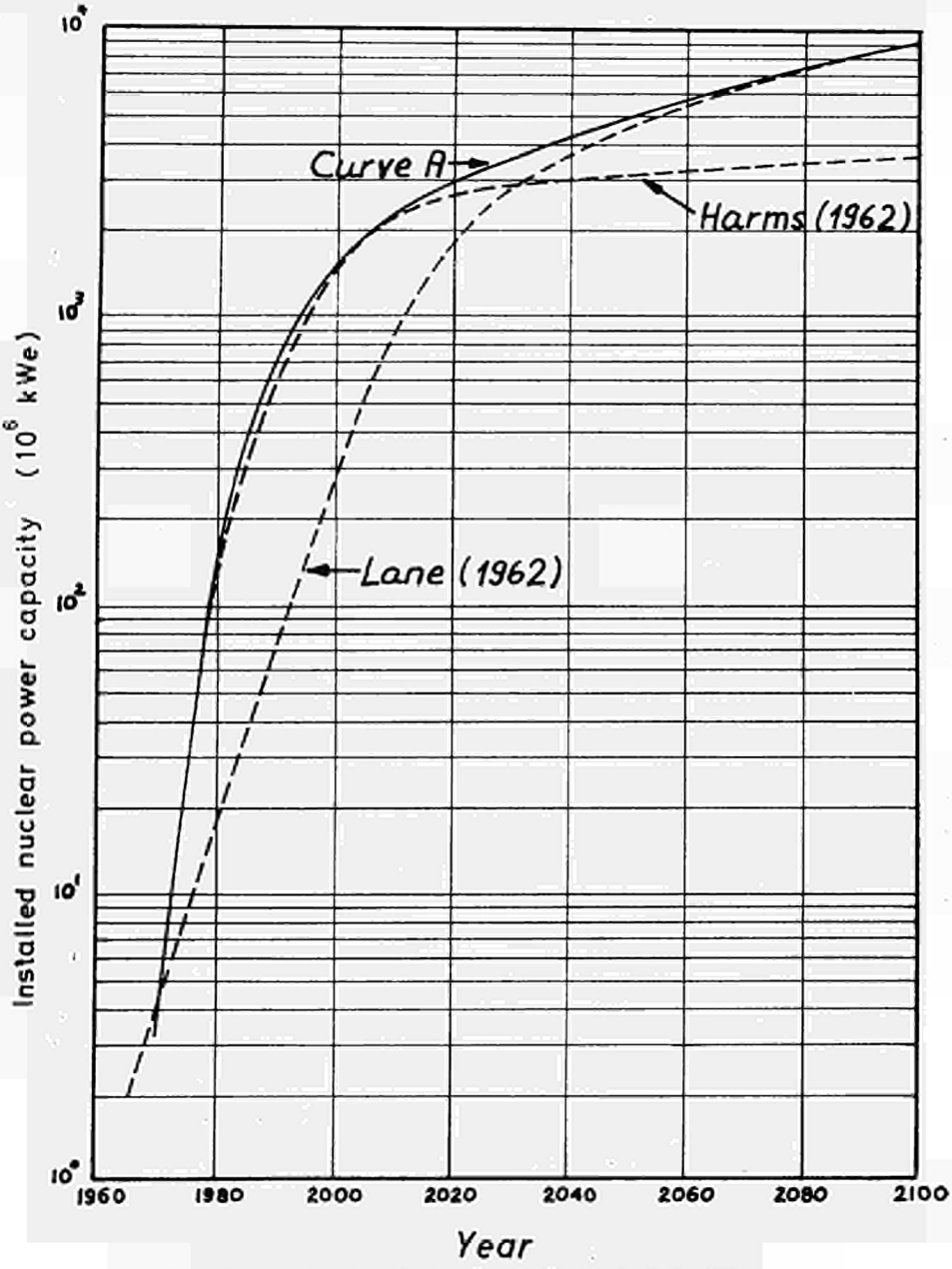

Fig. 1. — Estimated Nuclear Power Demand of the USA.

inherent differences. Latest estimates concerning future nuclear power demand are given by Harms and Lane. A basic load factor of 0.8 (according to a yearly utilization time of nearly 7,000 hours) will result the necessary installed capacity as a function of time (Fig. 1). For future estimates one should take into consideration that with a further decrease of costs the nuclear energy should not only serve as energy production but also for other purposes (e.g. desalting process). For that reason it will be prudent to use the maximal expected value for the necessary capacity requirements in order not to underestimate fuel requirements. In the case here, the envelope of the curves by Harms and Lane were chosen (curve A) and is described by the following analytical function :

$$\begin{aligned} P(t) &= 5 - 5{,}25\,(t - 1970) + 2{,}55\,(t - 1970)^2 \\ &\quad - 0{,}0275\,(t - 1970)^3 && t \leq 2000 \\ P(t) &= -730 + 71\,(t - 1970) && t \leq 2000 \end{aligned}$$

with

P = installed capacity covered by nuclear energy (10^6 kWe)
t = year

By application of bred fuel, the power capacity increases as follows :

$$\frac{dP^B}{dt} = aP(t) \tag{1}$$

P^B = installed power by inventory of bred fuel (10^6 KWe)
a = yearly variation ratio of fuel inventory

Factor a is dependent upon the physical properties of converters or breeders. For breeders, the equation

$$a = \frac{0.693}{Td} \tag{2}$$

is applied with Td being the doubling time, i.e. the time required to double fuel inventory.

The doubling time for converter reactors is not a reasonable physical size (it is negative). Therefore, the following definition is applied :

$$a\left[\frac{\text{Produced fuel amount per year in kg}}{\text{fuel inventory in kg}}\right] = (CR - 1) = \left[\frac{\text{Produced fuel amount in kg}}{\text{exhausted fuel amount in kg}}\right]$$

$$\rho\left[\frac{\text{kWe}}{\text{kg fuel}}\right] \cdot 1{,}2 \cdot 10^{-6}\left[\frac{\text{exhausted fuel amount per day in kg}}{\text{kWt}}\right]$$

$$\frac{1}{\eta}\left[\frac{\text{kWt}}{\text{kWe}}\right] \cdot 365\left[\frac{\text{days}}{\text{year}}\right]$$

$$\text{or} \quad a = 0.438(\text{CR} - 1)\frac{\rho}{\eta} \cdot 10^{-3} \tag{3}$$

with η = efficiency to change from thermal into electric power $\left(\frac{\text{kWe}}{\text{kWt}}\right)$

ρ = specific power $\left[\frac{\text{kWe}}{\text{kg fuel}}\right]$

In calculating the fuel amount expended for 1 kWt per day, differences arise of nearly 10 % because the energy per fission is not exactly known. Since the calculations here were done with the upper limit it seems not necessary to take into additional consideration the losses caused by fabrication and reprocessing, for they run up to only 2 %.

Definition for a for breeders and converters are equivalent, i.e. both equations give an equal result for equal physical dates. However, it should be considered that a for converters is a negative value. The integration of equation (1) results capacity P^{B} which is bred until year ϑ.

$$\text{P}^{\text{B}} = \int_0^{\vartheta} a \cdot \text{P}(t)dt \tag{4}$$

This capacity is not sufficient to cover the total requirements; difference P^{A} has to be reached by mining of fission material.

$$\text{P}^{\text{A}} = \text{P}(t) - \text{P}^{\text{B}} \tag{5}$$

Value P^{B} on the one hand depends upon the various parameters, on the other hand upon the model constructed to change converters into breeders. The four models given have a very simple mathematical structure; however, they bear a differential practical importance.

1. Constant Conversion Rate or Constant Doubling Time.

For the model here it was accepted that the conversion ratio or doubling time do not vary within the considered time. In order to work under these assumptions with low-cost reserves - based on the following calculation - it would be necessary to start with breeders in 1970. However, since one cannot expect technology will enable an economic application of breeders at that time, this model appears nearly unrealistic and will not be further discussed.

2. Step change from converters into breeders; after change-over, no converters will exist.

Also this model appears unrealistic since it will be impossible to find out inherent technological improvements to change all existing converters into breeders. From the economic view point, the possibility to close down at a certain time all converters and replace by breeders seems less reasonable.

3. After change-over all new plants will be breeders; converters will slowly come to an end.

This model seems to be in agreement with the future development. On the basis of a converter lifetime of 30 years, the equation will be as follows :

$$P_K\ (t) = P_K\ (T) - P_K\,(t - 30) \qquad t \geqq T$$
$$P_K\ (t) = 0 \qquad t \geqq T + 30$$

$P_K(t)$ = installed power in converters at time t
T = year of change-over from converters to breeders

For that case, equation (1) comes into

$$\frac{dP_K{}^B}{dt} = a(P_K(T) - P_K(t - 30)) \; ; \; t \geqslant T \tag{6}$$

Under consideration of the fuel exhausted in converters before change-over to breeders, the integration of this equation will be as follows :

$$P_K{}^B = aP_K(T)\,(t - T) - a\int_T^t P_K(t - 30)dt + a\int_0^T P_K(t)dt \tag{7}$$

Here, the following amount should be reached by mining to cover the total nuclear power demand :

$$P^A = P(t) - P_B{}^B - P^B{}_K \tag{8}$$

$P_B{}^B$ = capacity bred in breeders
$P_K{}^B$ = capacity bred in converters

4. Linear increase of conversion ratio.

In this case the conversion ratio is supposed to increase linearly from a starting value to a value of CR = 1 in the year of change-over. In the following time, only breeders with a certain doubling time will be built. The starting value was chosen as CR = 0.6 in 1970. By this, the different years of change-over give the following functions for an increase of the conversion ratio :

1980	CR = 0.6 + 0.04	$(t - 1970)$
1990	CR = 0.6 + 0.02	$(t - 1970)$
2000	CR = 0.6 + 0.0133	$(t - 1970)$
2010	CR = 0.6 + 0.01	$(t - 1970)$

5. — Results

The calculation results of models 3 and 4 are given in the following diagrams. Fig. 2-4 show the total necessary fuel requirement as a function of the year of change-over from converters to breeders with various conversion ratios as parameters and for different specific powers. Fig. 5-13 show the total necessary fuel requirement as a function of time for different years of change-over. Fig. 5-9 show the influence

of the conversion ratio on the fuel requirement at a doubling time of Td = 25 years and a specific power = $1 \frac{\text{MWe}}{\text{kgU}_5}$. (The fission material consists in the beginning of U^{235}, later additional isotopes of U^{233} and Pu^{239} are to be contained in "U_5"). Fig. 10-17 show the influence of doubling time and specific power.

The points plotted in Fig. 2-4 are in accordance with the maxima of the curves in Fig. 9-17. In these figures the available reserves are listed. They are expressed as

A = 270,000 short tons U_3O_8, sure available reserves of category < 10$ per lb U_3O_8 according to Faulkner and McVey;

B = 520,000 short tons U_3O_8, total reserves of category < 10$ per *lb* U_3O_8 according to Faulkner and McVey;

C = 1,220,000 short tons U_3O_8, total reserves with possibility to be mined at maximal 30$ per *lb* U_3O_8 according to Faulkner and McVey;

D = 2,530,000 short tons U_3O_8, total reserves of category < 10$ per lb U_3O_8 according to McKelvey.

The intersection points of the requirement curves with the lines of the available reserves result the time of the change-over from converters to breeders, for which the referring reserves are sufficient to cover the energy requirement. Fig. 18-27 give same results for a linear increase of the conversion ratio (model 4).

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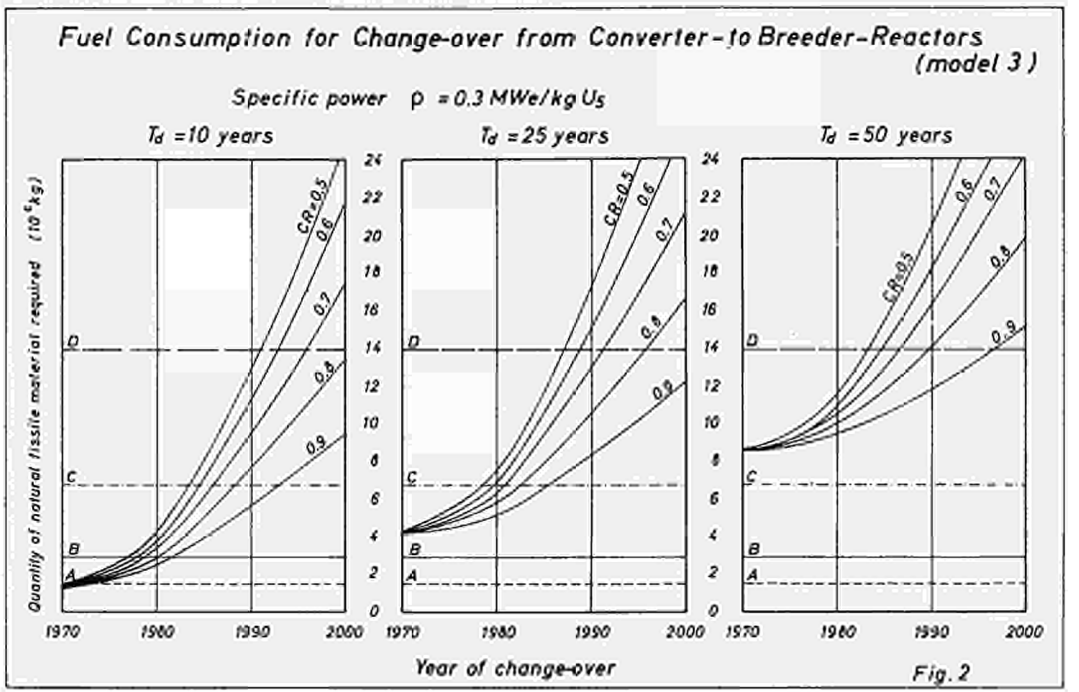
Fuel Consumption for Change-over from Converter-to Breeder-Reactors
(model 3)
Specific power ρ = 0.3 MWe/kg U5
Td = 10 years
Td = 25 years
Td = 50 years
Quantity of natural fissile material required (10⁶ kg)
CR=0.5
0.6
0.7
0.8
0.9
A
B
C
D
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
Year of change-over
Fig. 2

FIG. 2.

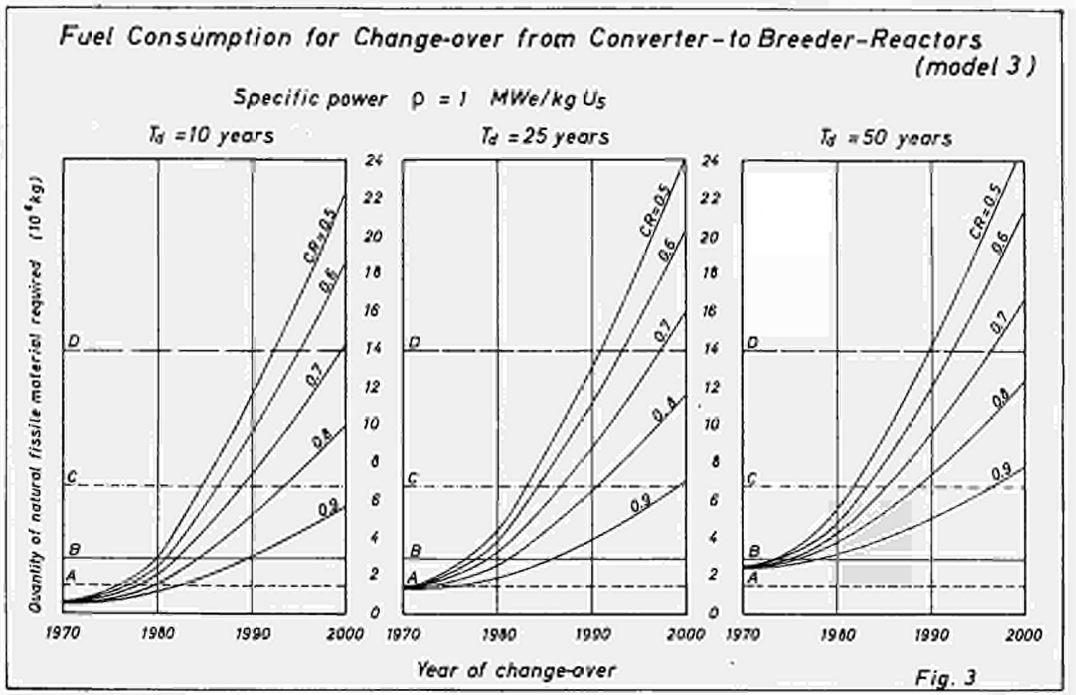
Fuel Consumption for Change-over from Converter-to Breeder-Reactors
(model 3)
Specific power ρ = 1 MWe/kg U5
Td = 10 years
Td = 25 years
Td = 50 years
Quantity of natural fissile material required (10⁶ kg)
CR=0.5
0.6
0.7
0.8
0.9
0.4
A
B
C
D
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
Year of change-over
Fig. 3

FIG. 3.

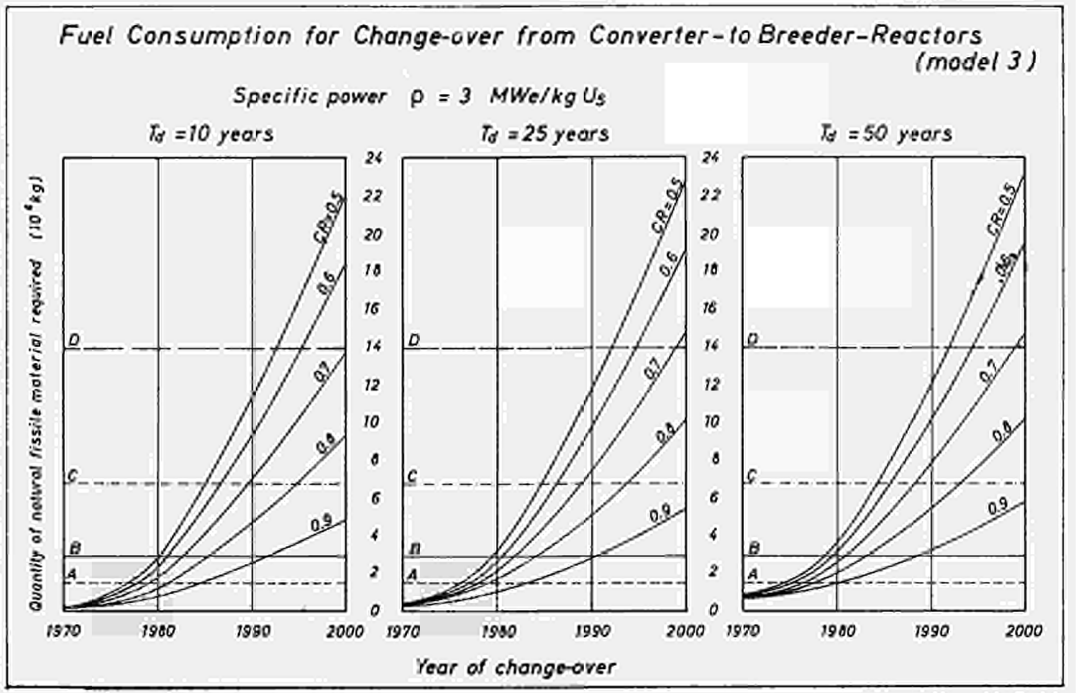
Fuel Consumption for Change-over from Converter-to Breeder-Reactors
(model 3)
Specific power p = 3 MWe/kg Us
Td =10 years
Td =25 years
Td =50 years
Quantity of natural fissile material required (10⁶ kg)
CR=0.5
0.6
0.7
0.8
0.9
D
C
B
A
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
Year of change-over

FIG. 4.

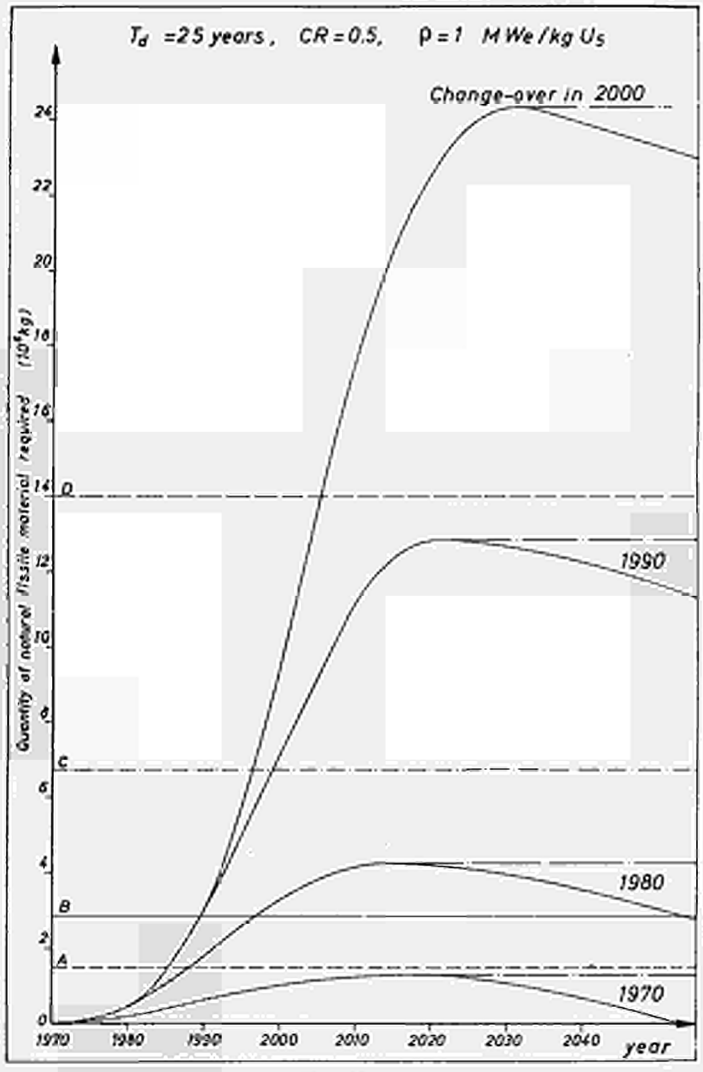
T_d =25 years, CR=0.5, ρ=1 MWe/kg U_S
Change-over in 2000
1990
1980
1970
Quantity of natural fissile material required (10⁴kg)
year
A
B
C
D
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
2010
2020
2030
2040

FIG. 5.

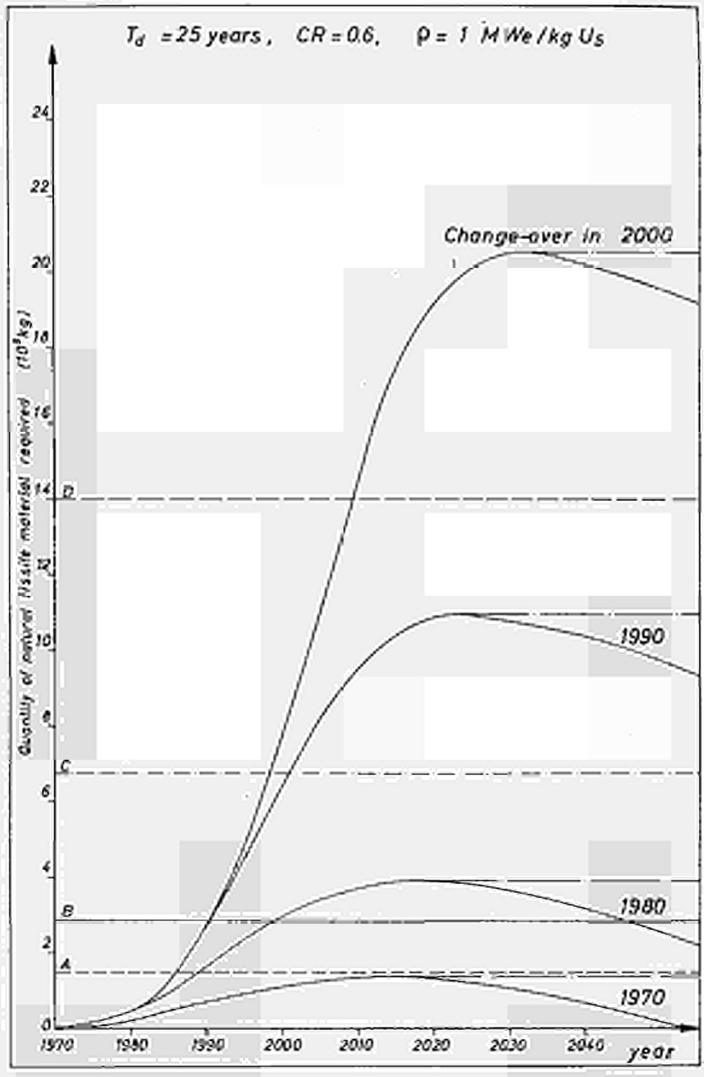
T_d = 25 years, CR = 0.6, ρ = 1 MWe/kg U_S
Change-over in 2000
1990
1980
1970
A
B
C
D
Quantity of natural fissile material required (10³kg)
year
1970
1980
1990
2000
2010
2020
2030
2040
0
2
4
6
8
10
12
14
16
18
20
22
24

FIG. 6.

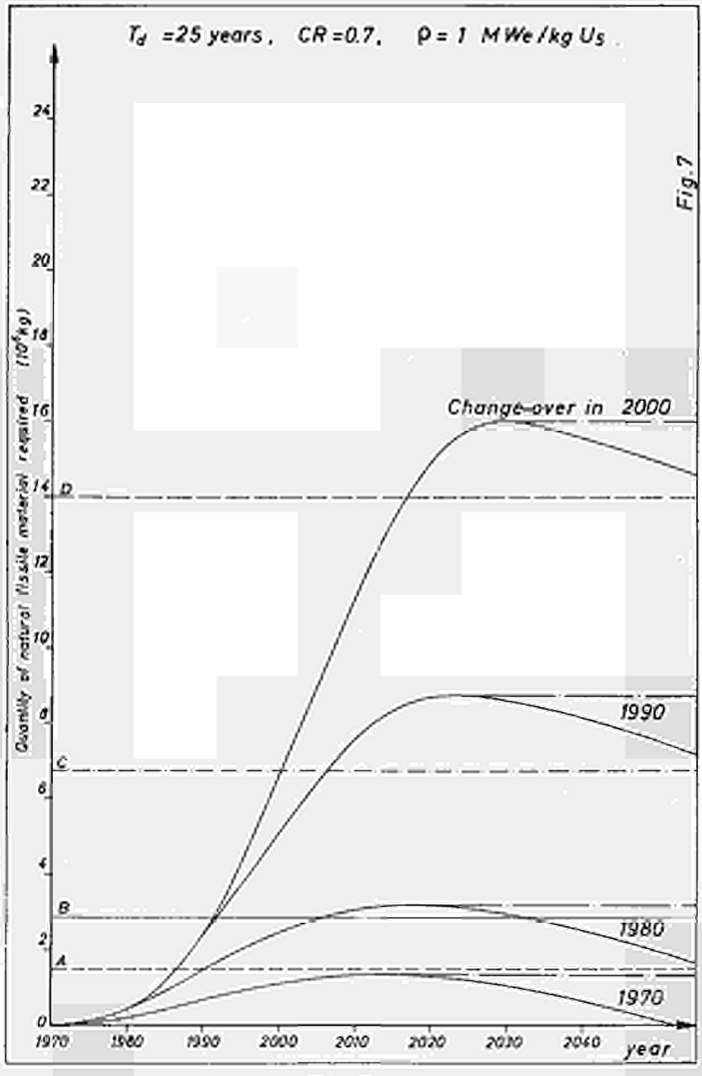
T_d =25 years, CR=0.7, ρ = 1 MWe/kg Us
Fig.7
Quantity of natural fissile material required (10⁶kg)
Change-over in 2000
1990
1980
1970
A
B
C
D
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 7.

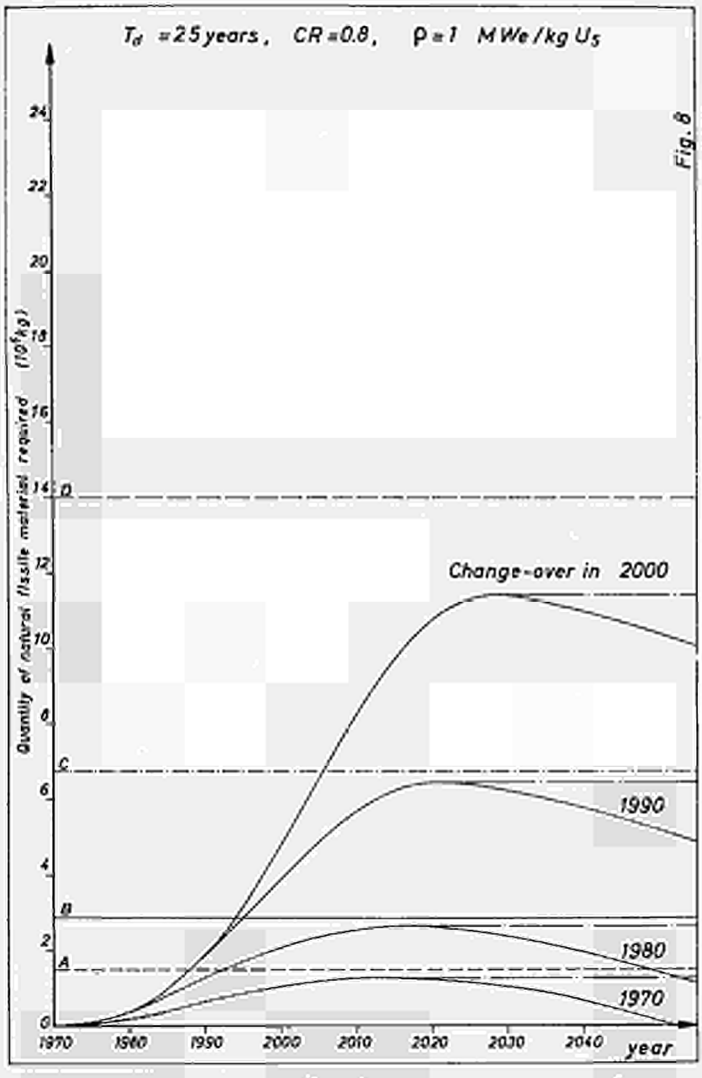
T_d = 25 years, CR = 0.8, P = 1 MWe/kg U_5
Fig. 8
Quantity of natural fissile material required (10^6 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 8.

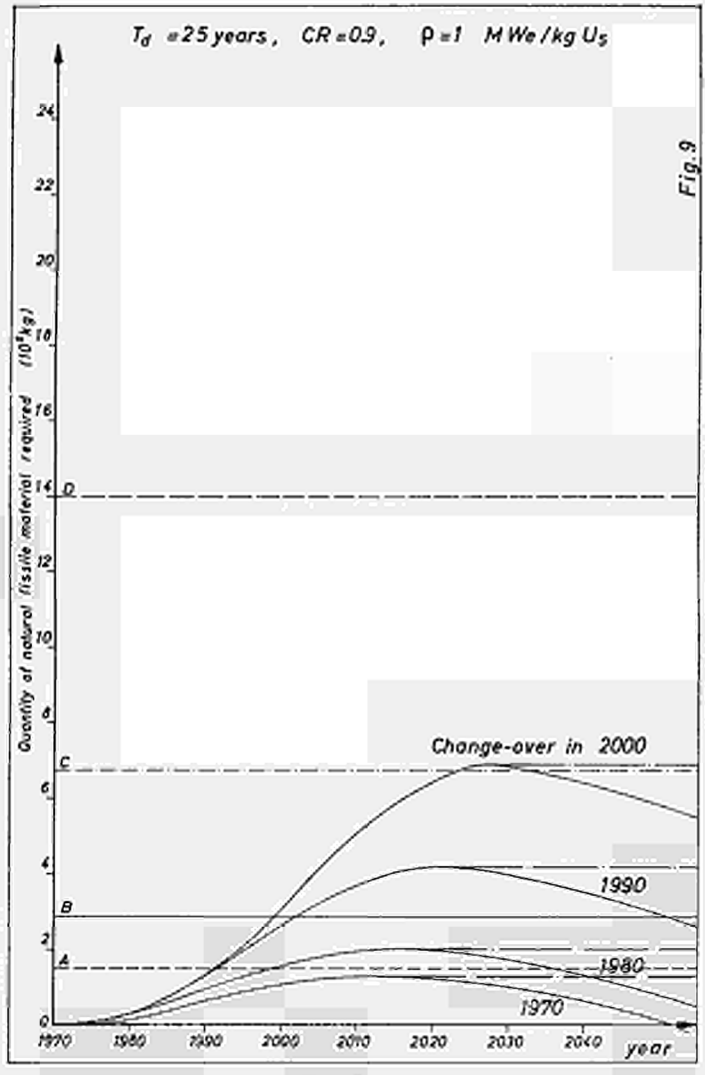
T_d = 25 years, CR = 0.9, ρ = 1 MWe/kg U_S
Fig. 9
Quantity of natural fissile material required (10^6 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 9.

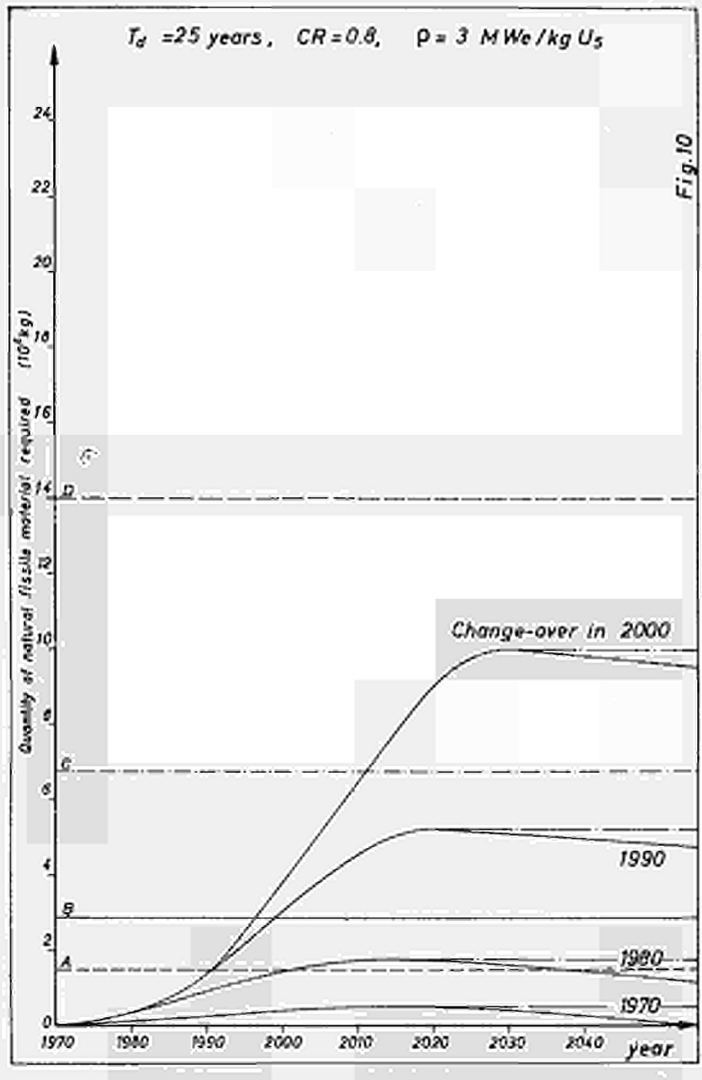
T_d =25 years, CR=0.8, ρ = 3 MWe/kg U_5
Fig.10
Quantity of natural fissile material required (10^6kg)
Change-over in 2000
1990
1980
1970
A
B
C
D
year
1970
1980
1990
2000
2010
2020
2030
2040

FIG. 10.

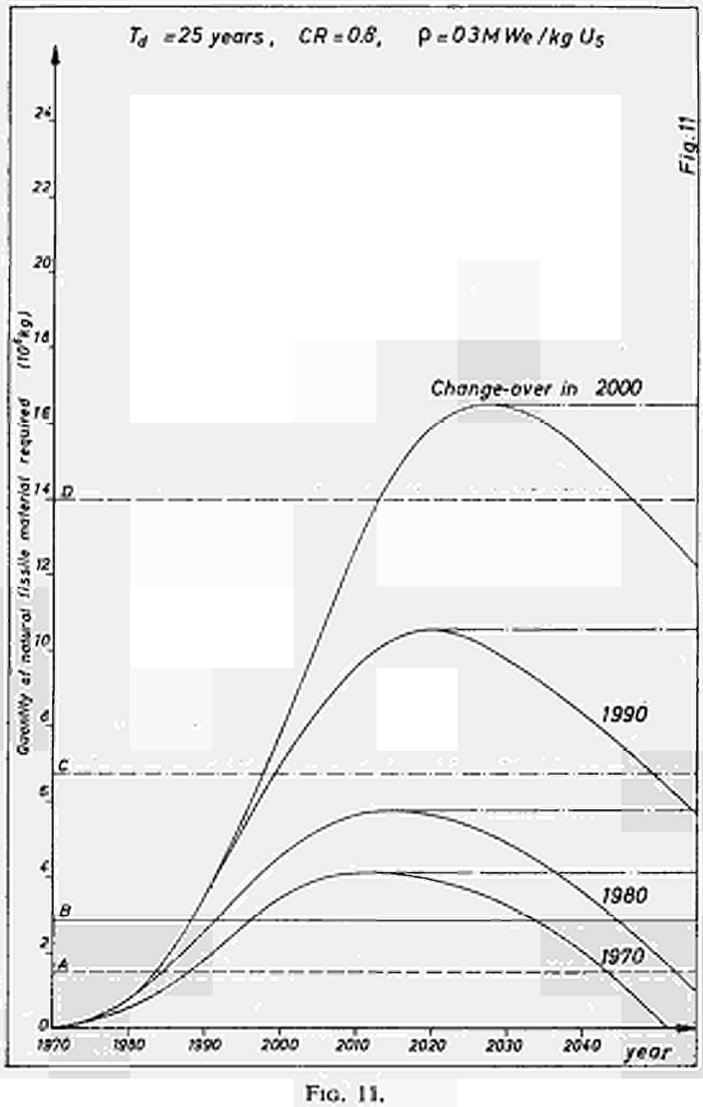
T_d = 25 years, CR = 0.8, ρ = 0.3 MWe/kg U_5
Fig. 11
Quantity of natural fissile material required (10^6 kg)
Change-over in 2000
1990
1980
1970
D
C
B
A
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 11.

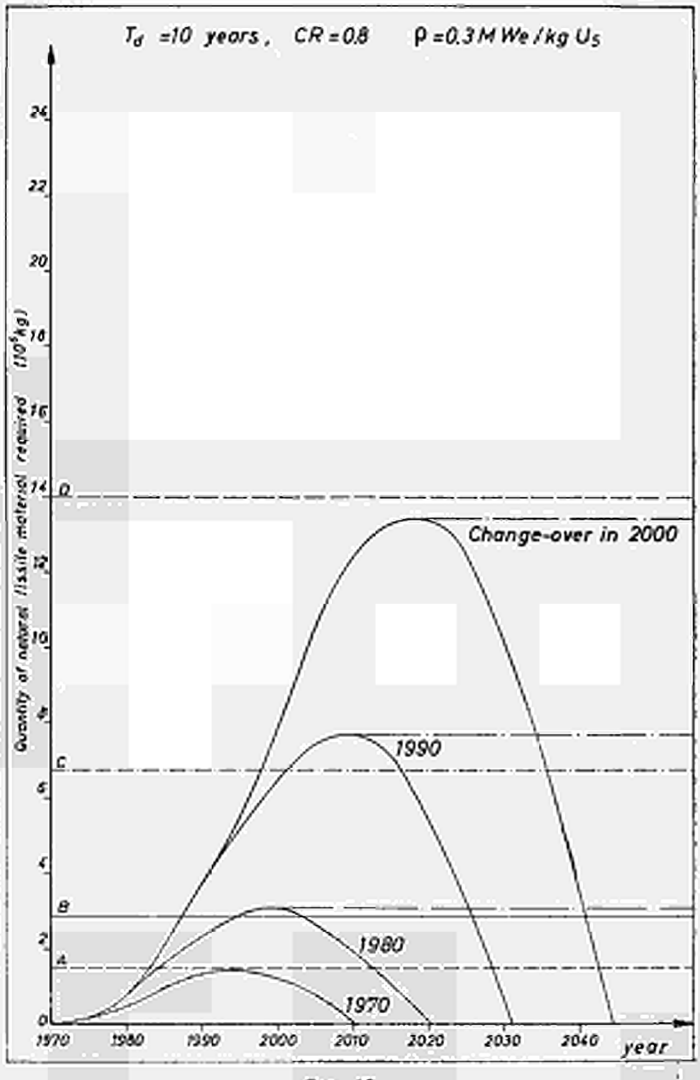
T_d =10 years, CR=0.8 ρ=0.3 MWe/kg U_5
Quantity of natural fissile material required (10^5 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 12.

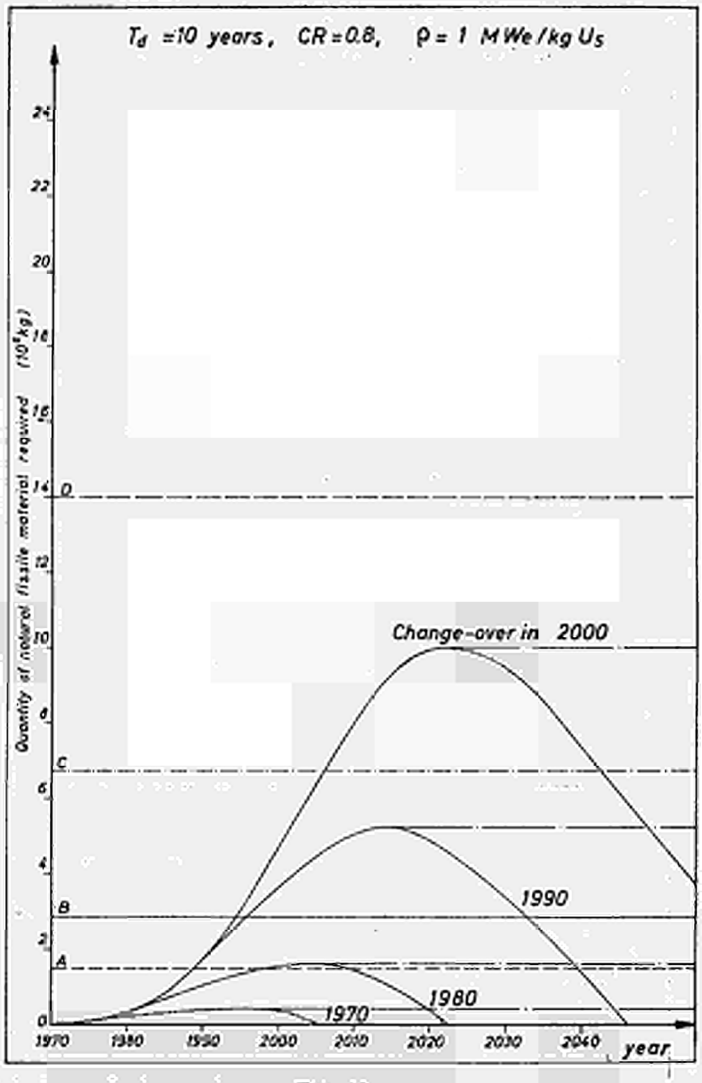
T_d =10 years, CR=0.8, ρ = 1 MWe/kg U_S
Quantity of natural fissile material required (10^6kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 13.

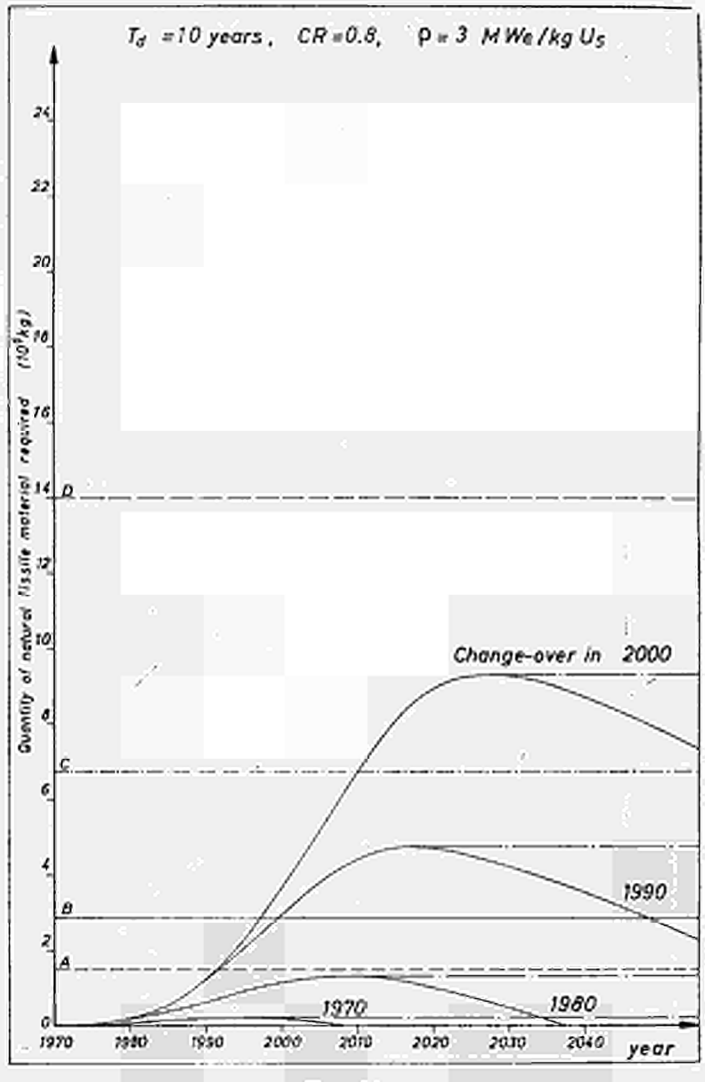
T_d =10 years, CR=0.8, ρ = 3 MWe/kg U_S
Quantity of natural fissile material required (10^3kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1970
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 14.

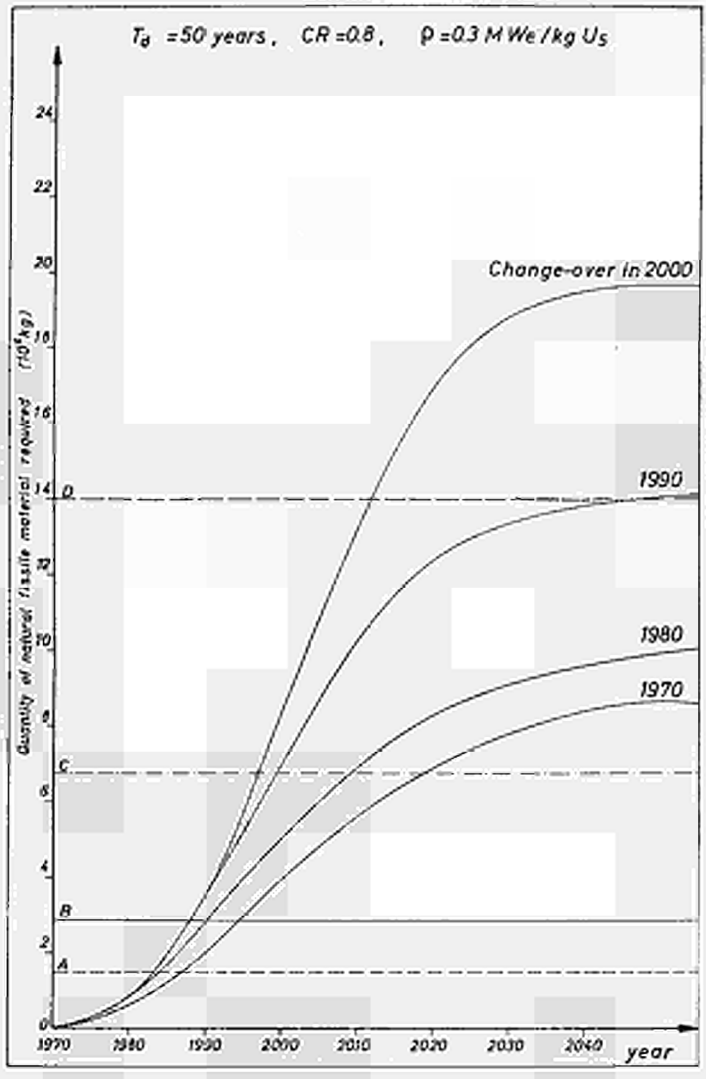
T_d =50 years, CR=0.8, ρ=0.3 MWe/kg U_S
Change-over in 2000
1990
1980
1970
D
C
B
A
Quantity of natural fissile material required (10^4kg)
0
2
4
6
8
10
12
14
16
18
20
22
24
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 15.

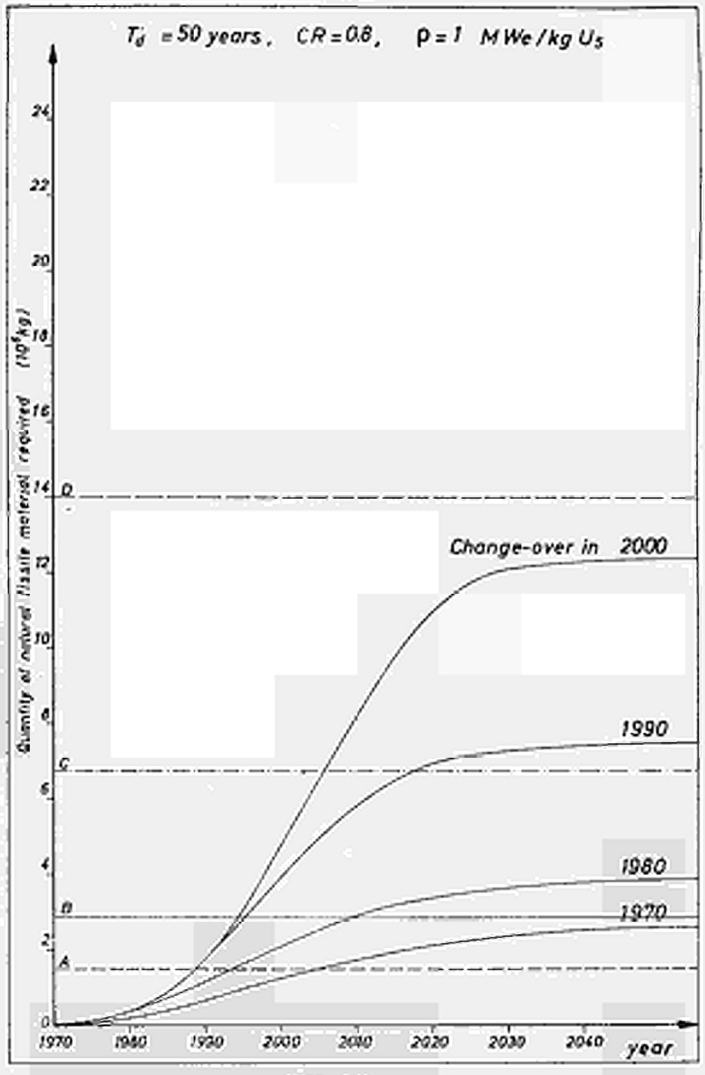
T_d = 50 years, CR = 0.8, ρ = 1 MWe/kg U_5
Quantity of natural Uranium material required (10^6kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 16.

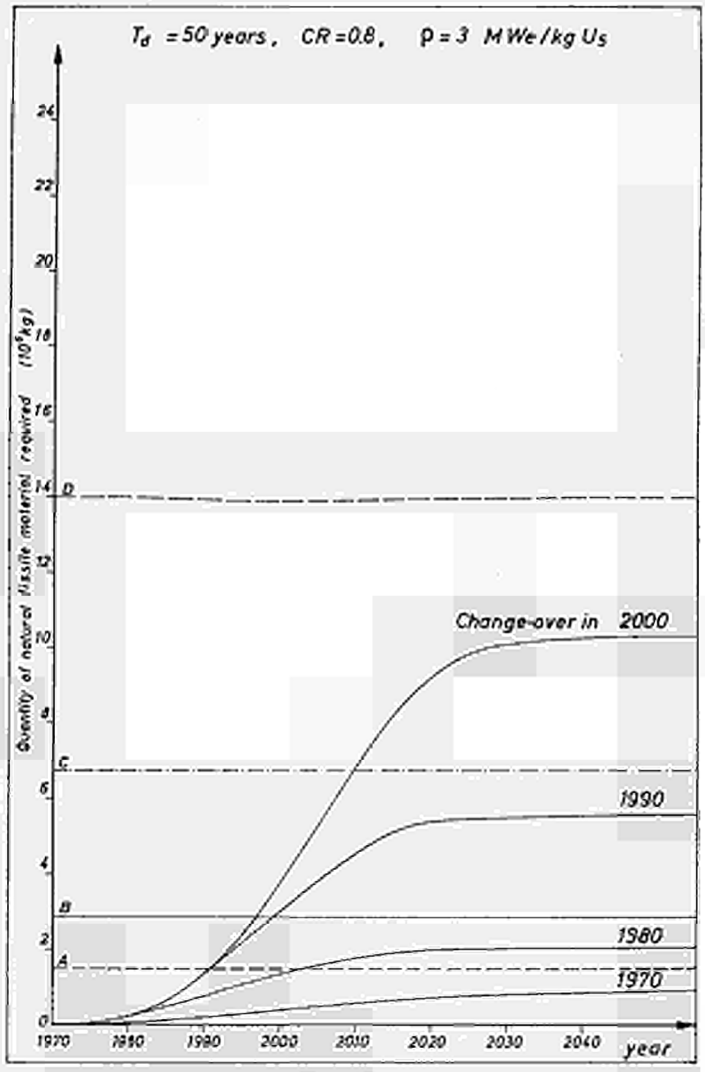
T_d = 50 years, CR = 0.8, p = 3 MWe/kg Us
Quantity of natural fissile material required (10^6kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2000
1990
1980
1970
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 17.

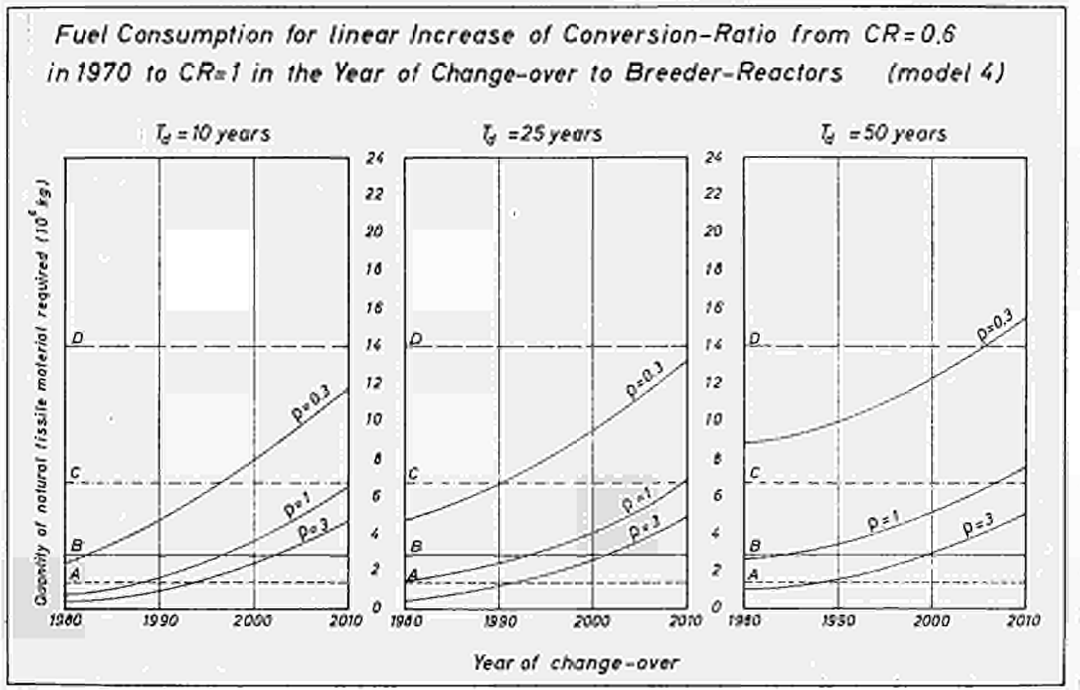
Fuel Consumption for linear Increase of Conversion-Ratio from CR=0.6 in 1970 to CR=1 in the Year of Change-over to Breeder-Reactors (model 4)
T_u = 10 years
T_u = 25 years
T_u = 50 years
Quantity of natural fissile material required (10^6 kg)
Year of change-over
A
B
C
D
ρ=0.3
ρ=1
ρ=3
1980
1990
2000
2010
0
2
4
6
8
10
12
14
16
18
20
22
24

FIG. 18.

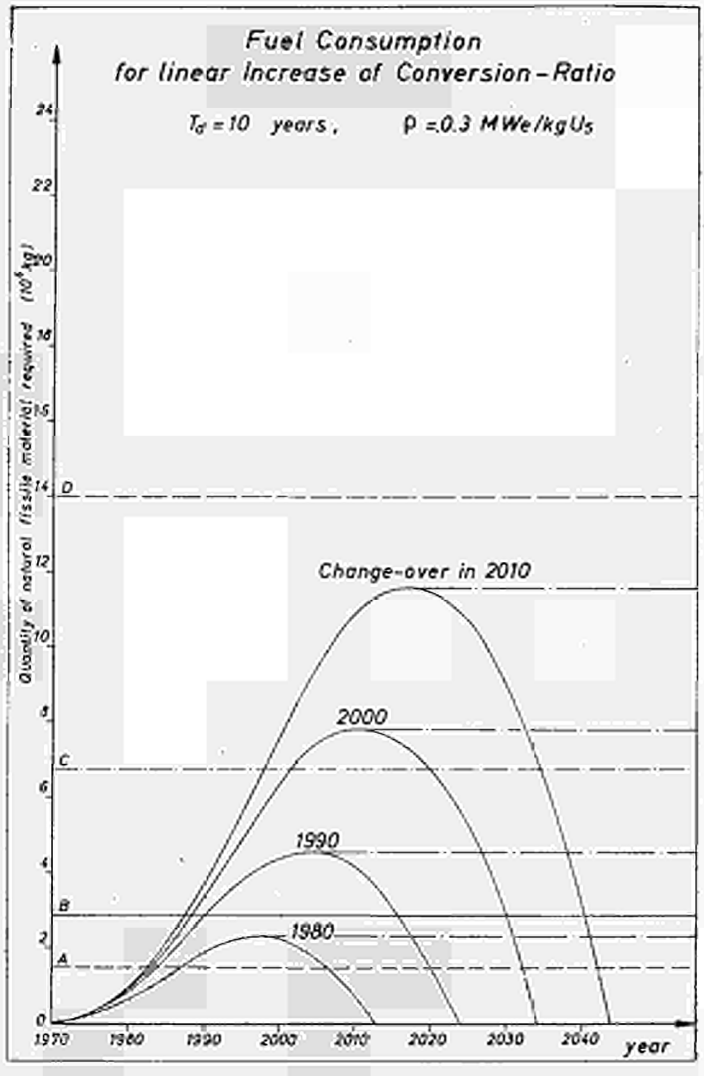
Fuel Consumption
for linear Increase of Conversion-Ratio
$T_d = 10$ years, $\rho = 0.3$ MWe/kg U_S
Quantity of natural fissile material required (10^3 kg)
Change-over in 2010
2000
1990
1980
A
B
C
D
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 19.

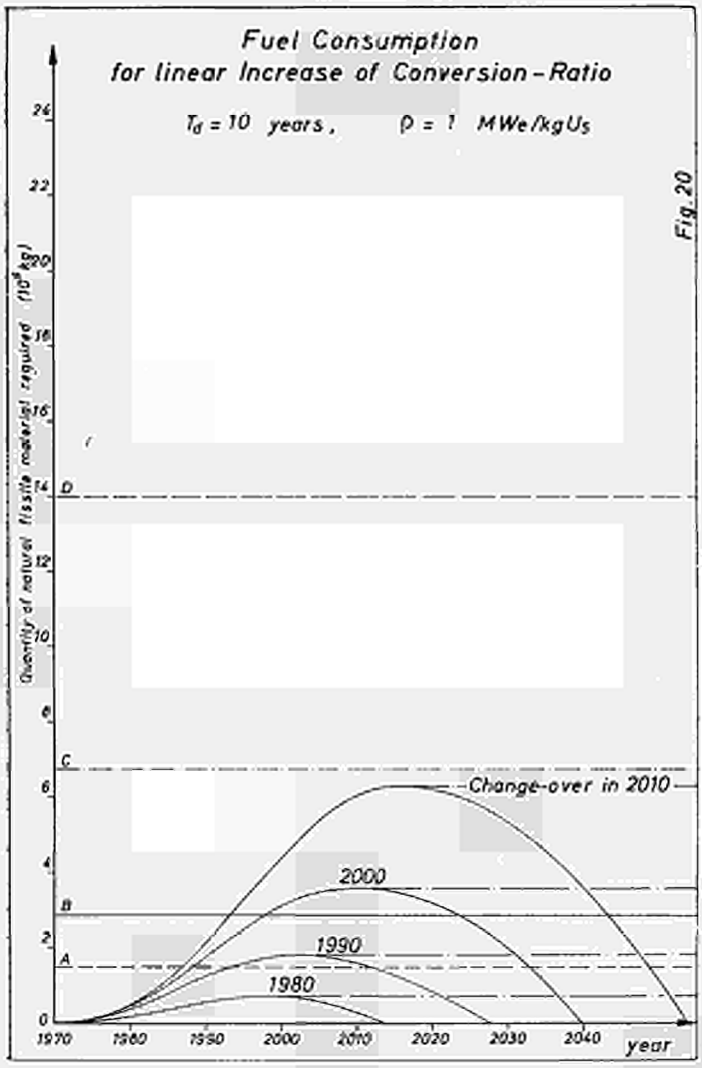
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 10 years, P = 1 MWe/kgU$_S$
Fig. 20
Quantity of natural fissile material required (10^3 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1960
1950
2000
2010
2020
2030
2040
year

FIG. 20.

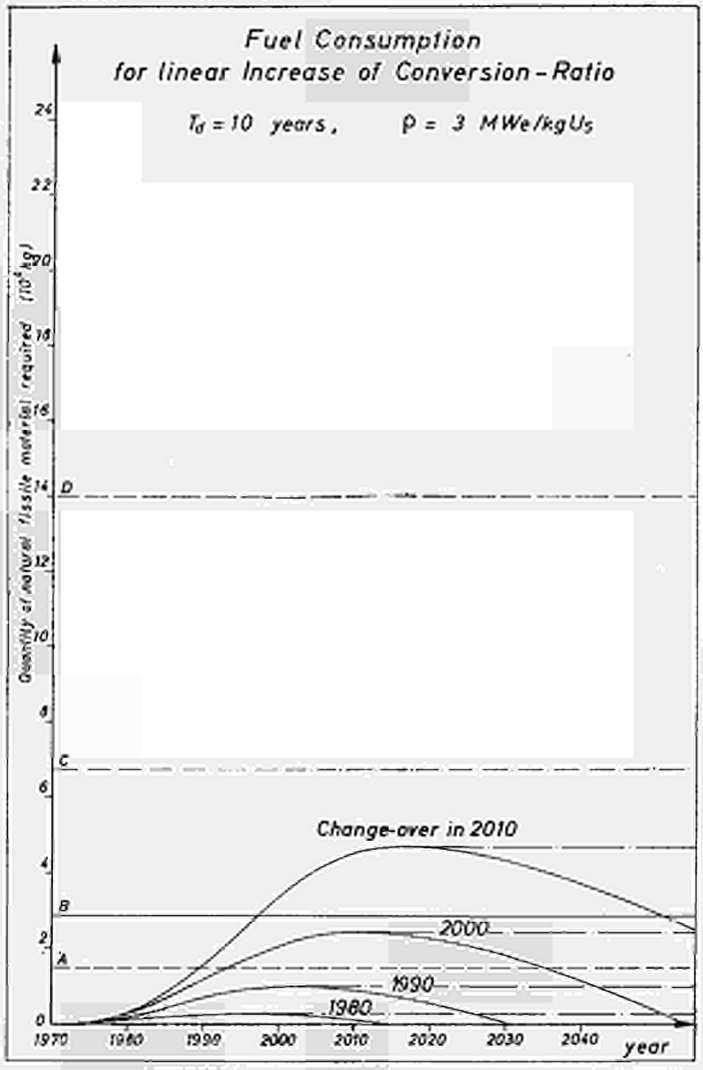
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 10 years, ρ = 3 MWe/kgU_S
Quantity of natural fissile material required (10^4 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 21.

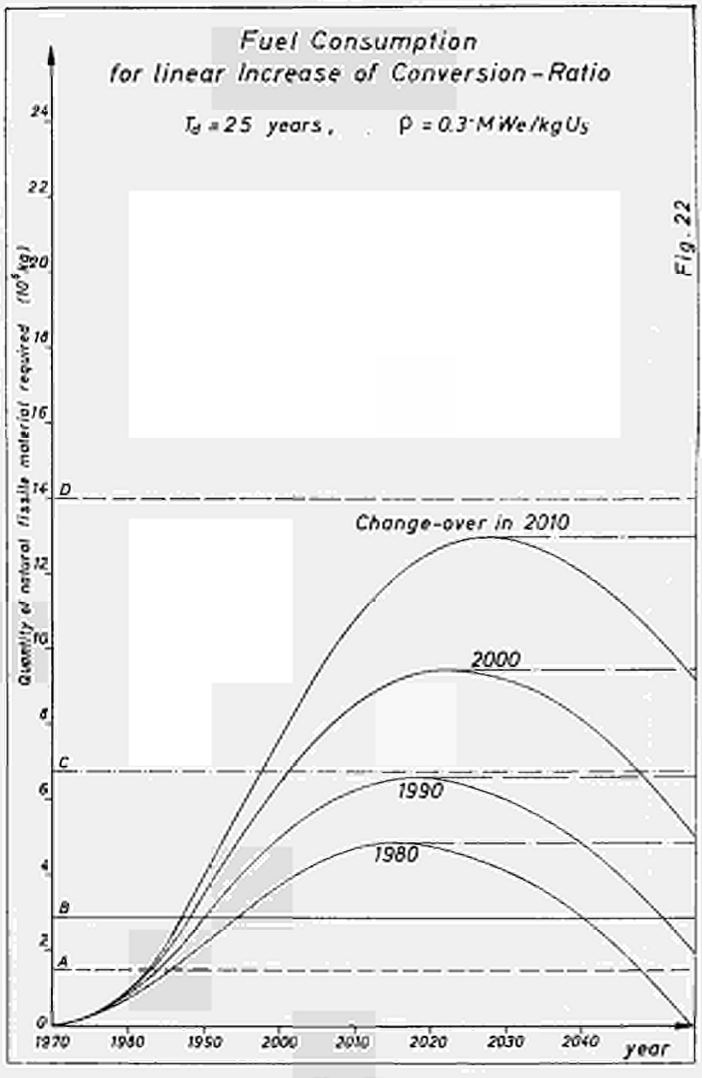
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 25 years, ρ = 0.3 MWe/kg U_S
Fig. 22
Quantity of natural fissile material required (10^3 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 22.

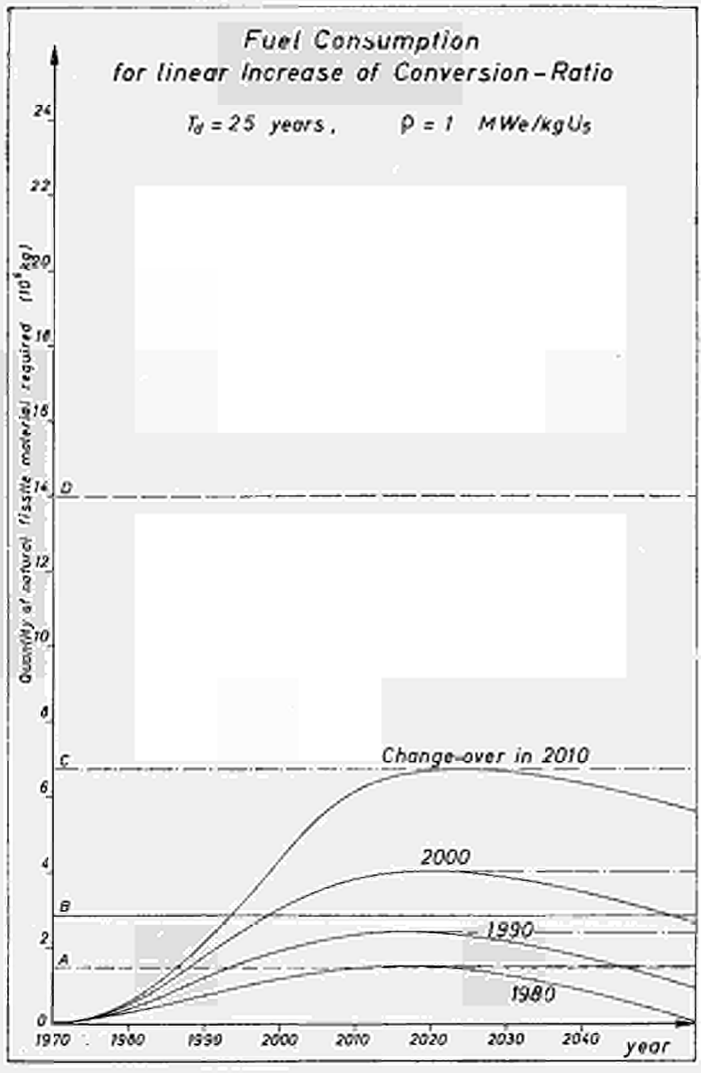
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 25 years, ρ = 1 MWe/kg U_S
Quantity of natural fissile material required (10^6 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 23.

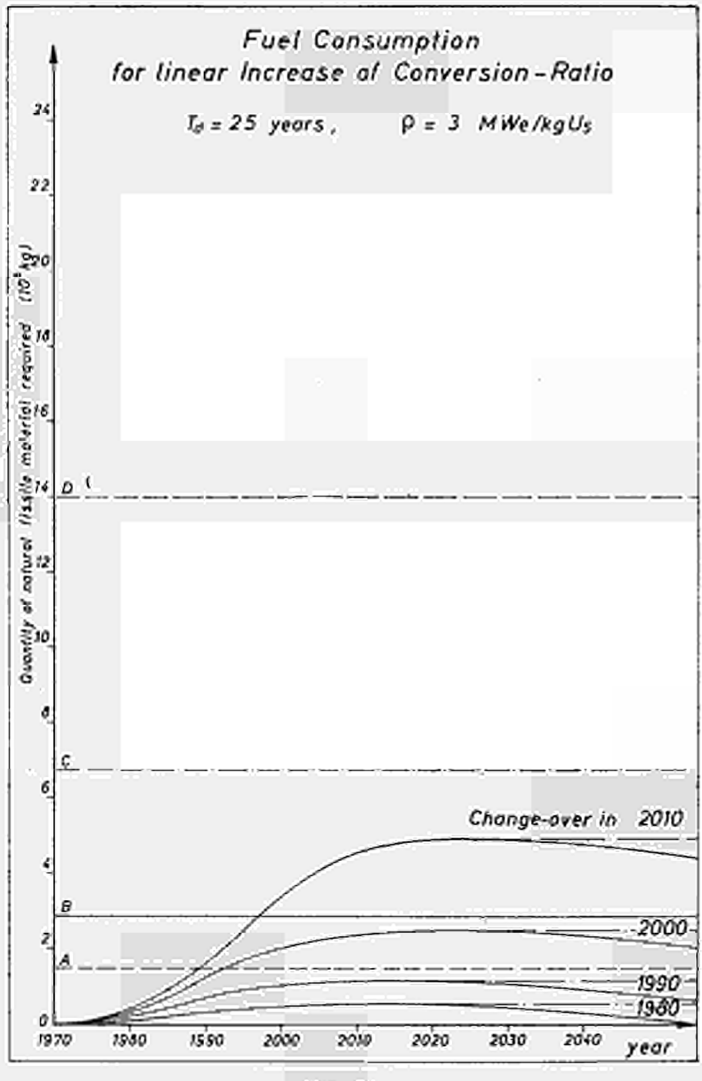
Fuel Consumption
for linear Increase of Conversion-Ratio
T_v = 25 years, ρ = 3 MWe/kgU$_5$
Quantity of natural fissile material required (10^3 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1950
1550
2000
2010
2020
2030
2040
year

FIG. 24.

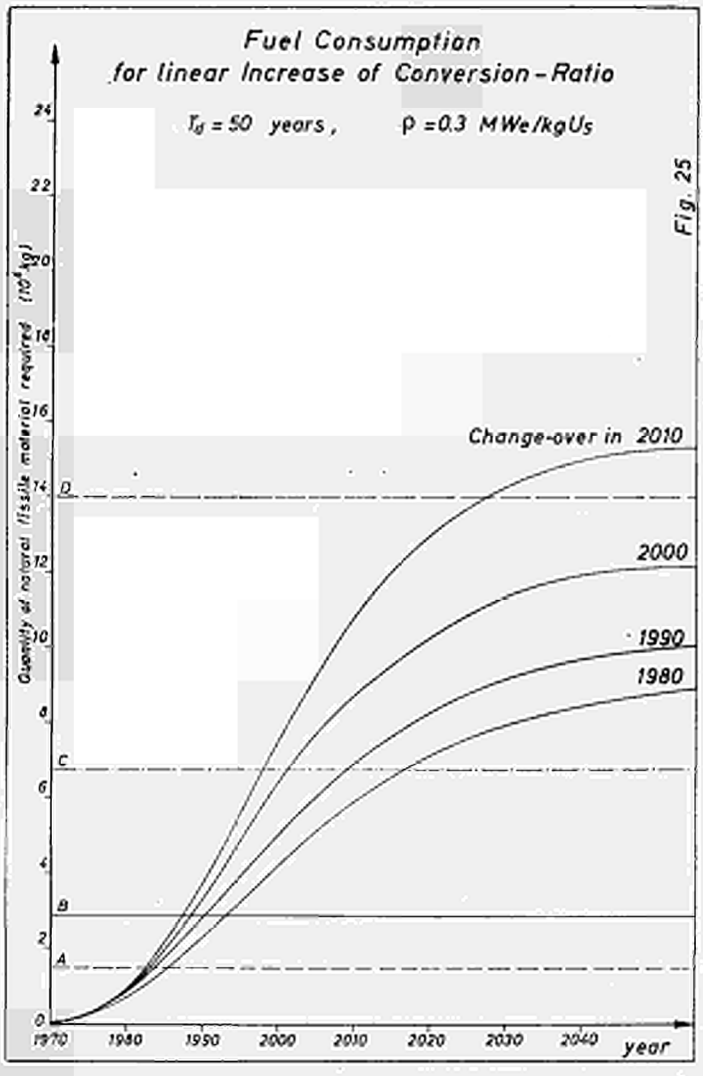
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 50 years, ρ = 0.3 MWe/kgUs
Fig. 25
Quantity of natural fissile material required (10⁴ kg)
Change-over in 2010
2000
1990
1980
D
C
B
A
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 25.

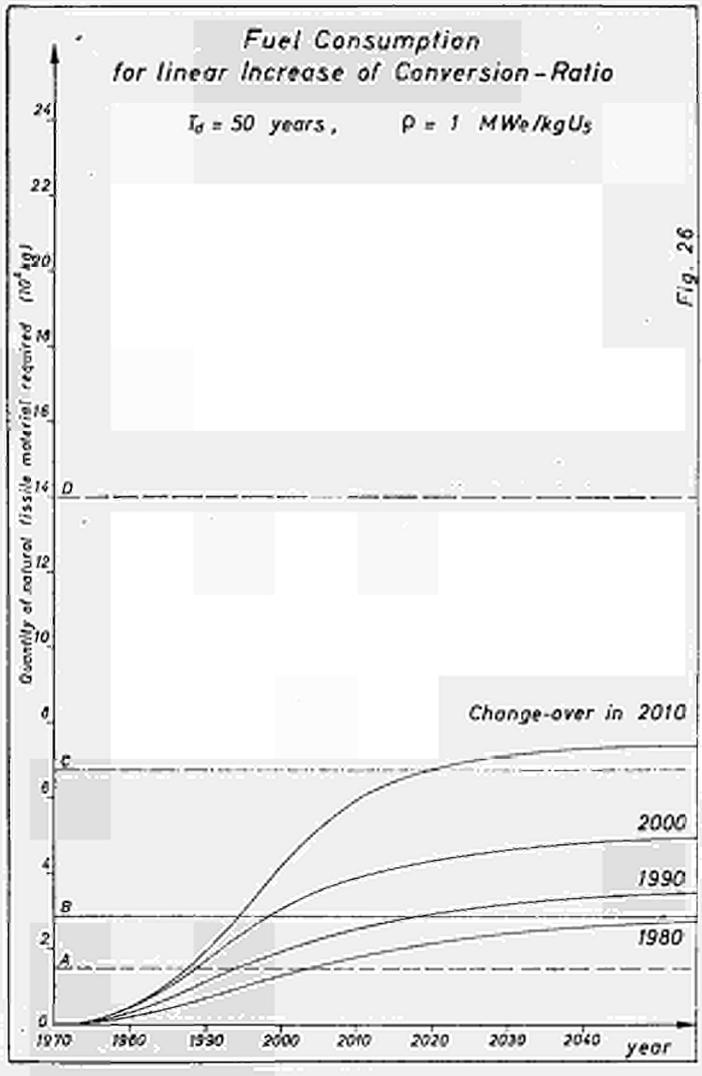
Fuel Consumption
for linear Increase of Conversion-Ratio
T_d = 50 years, ρ = 1 MWe/kgUs
Fig. 26
Quantity of natural fissile material required (10^4 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 26.

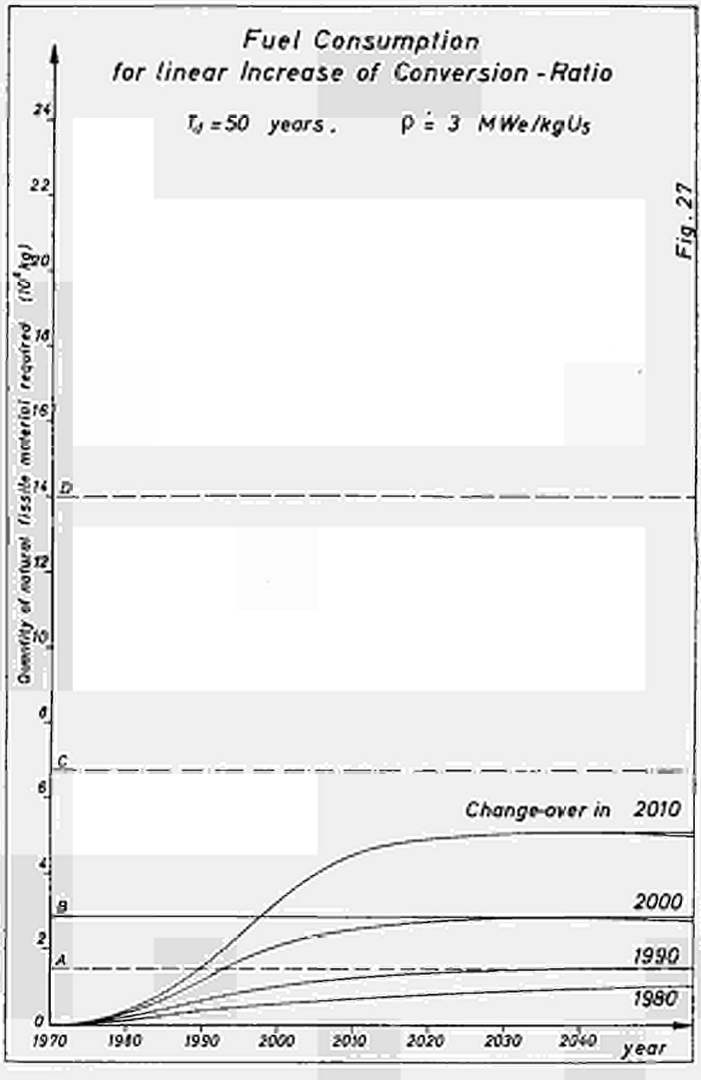
Fuel Consumption
for linear Increase of Conversion - Ratio
T_u = 50 years. $\dot{p}$ = 3 MWe/kgU$_5$
Fig. 27
Quantity of natural fissile material required (10^4 kg)
24
22
20
18
16
14
12
10
8
6
4
2
0
D
C
B
A
Change-over in 2010
2000
1990
1980
1970
1980
1990
2000
2010
2020
2030
2040
year

FIG. 27.

HTGR FUEL CYCLE ASSESSMENT STUDIES [1]

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General Atomic Division of General Dynamics
San Diego, California, U.S.A.

ABSTRACT

The probable nuclear resource requirements and the projected fuel cycle costs of the High Temperature Gas-Cooled Reactor (HTGR) have been evaluated relative to other thermal spectrum reactors, including other advanced converters and fast spectrum breeder reactors. To account for the evolution of reactor types, symbiotic systems of thermal and fast spectrum reactors have also been included.

To utilize efficiently the available nuclear resources in a rapidly expanding power economy, with the restriction of competitive power costs, a reactor concept must minimize both (*a*) the fuel inventory required to start up additional reactors and (*b*) the net fuel consumed to produce energy. To meet these demands, a reactor concept must perform well in four areas, i.e. it must have :

1. a high thermal efficiency
2. a high specific power
3. a high conversion ratio
4. a reasonably long fuel irradiation time relative to the time spent by the fuel outside the reactor core.

Advanced converter and fast reactor concepts must utilize U^{233} or Pu^{239} to achieve these four characteristics. Since both U^{233} and Pu^{239} must be bred in reactors using U^{235}, a complete evaluation of the resource requirements and power costs of an advanced reactor system in an expanding power economy involves a dynamic evaluation considering the reactors producing the desired fuel (i.e. converter or feeder reactors) as well as the reactors utilizing the bred fuel (i.e. recycle or breeder reactors). Such a symbiotic analysis shows that an HTGR-HTGR feeder-breeder system has about the same uranium requirements as a thermal-fast U/Pu symbiotic system over the course of the next half century.

The fast breeder reactors must achieve conversion ratios in excess of 1.3 and specific powers greater than 1,000 KW/kg to compete with the nuclear fuel requirements and the fuel cycle cost of the HTGR-HTGR system. Despite the importance of conserving nuclear resources, the criterion for acceptance of a reactor concept will be economic performance. Since low costs ores will probably be exhausted within the next 30 to 50 years depending on the types of reactors used and their performance characteristics, fuel cycle costs at higher ore costs must be carefully considered in the choice of reactor concepts for the long-range future. Because of the high specific power, high conversion ratio, and high thermal efficiency of the HTGR, its fuel cycle cost is relatively insensitive to ore cost. The economic potential for the HTGR appears to be better than that of other advanced converter concepts. The ability of the HTGR to compete with fast breeder concepts will depend on the ultimate performance that can be realized in the very long range for each of the concepts within the limits of technology and safety.

[1] Work supported in part under U.S. Atomic Energy Commission Contract AT(04-3)-167 and in part under privately supported work at General Atomic, Report GA-6146, May 1965.

1. — Introduction

The AEC Civilian Nuclear Power Report to the President, which was published in 1962 [1], presents an excellent analysis of the long-range importance of nuclear energy in the United States and outlines a proposed program to meet both the intermediate and the long-range objectives of our power generation industry. The report discusses the potential roles of both the advanced converter and the breeder reactors in meeting the over-all objectives of the program.

More than two years have elapsed since the 1962 report was prepared, and additional studies on both the advanced converter and the breeder reactor concepts make it possible to examine in somewhat greater detail the probable roles that these concepts will fill in the next few decades. The purpose of this paper is to indicate the potential role we anticipate for the advanced converter reactor based on recent work done at General Atomic. In order to assess the potential of the HTGR, we have examined the probable fuel utilization and projected fuel cycle economics of the HTGR relative to existing reactor concepts, to other advanced converters, to fast breeder reactors, and to combinations of reactor systems.

In the subsequent discussions, results of calculations and analysis will be presented that support the following conclusions :

1. A high conversion or breeding ratio, per se, does not assure minimum nuclear fuel requirements in a growing nuclear power economy.
2. Minimizing the fuel requirements in a power reactor complex does not, per se, assure the long-range effective utilization of nuclear resources.
3. The amount of uranium ore projected [1] to be available in the United States at prices less than $10 per pound is insufficient to support the expected energy requirements for the next fifty years almost independent of the types of reactors built.
4. Since a rapidly growing, large nuclear power industry will almost certainly require the use of more expensive ore, the most critical index for choosing an attractive reactor concept is the economic performance potential of the reactor relative to other energy-conversion systems when the price of uranium ore has risen to, say, $20 or $30 per pound. It is within this context that we must interpret the objective of maximum utilization of nuclear resources. Conservation of nuclear resources for its own sake is, therefore, not the overriding consideration in the maximum utilization of nuclear resources. It is generally true, however, that reactor concepts capable of generating economic power with relatively expensive ore are also concepts that use the uranium ore efficiently and therefore tend to conserve resources.
5. In order to meet the objectives of favorable utilization of nuclear resources under economically attractive conditions, it is found that a reactor concept should simultaneously have the following four characteristics :
   (*a*) A high thermodynamic efficiency.
   (*b*) A high conversion ratio.

(*c*) A high specific power.
(*d*) A reasonably long fuel irradiation time relative to the time spent by the fuel outside the reactor core.

6. When one judges reactor concepts on the basis of economic attractiveness under conditions of increasing ore prices, the potential for the HTGR appears to be better than that of other advanced converter concepts, and in many circumstances competitive with fast breeder reactors.

The basis for these conclusions will be developed in the succeeding discussion. Section 2 will review the electric power and energy forecasts for the next few decades and the estimated availability of uranium and thorium resources. Section 3 will examine the cumulative resource requirements of various reactor types. Section 4 will then look at the fuel cycle economics under conditions typified by the various reactor concepts. Section 5 will cover in somewhat closer detail the uranium commitments and fuel cycle economics associated with combinations of converter and recycle reactors including both near breeders and breeders.

2. — Nuclear power forecasts and resource requirements

The projected growth of the United States nuclear power generation capacity to the year 2020 A.D. as forecast by the AEC [1, 2] has been taken as the basis of the analysis to be described in this report. An interpretation of the nuclear electric generating capacity and doubling time projected by the AEC in 1962 was presented by Dietrich [3] in his paper on efficient utilization of nuclear fuels. In this growth curve, a linear increase of generating capacity after the year 2000 A.D. was assumed. This has the effect of increasing the doubling time from six years to thirty years over a time interval of thirty years. In view of the fact that the doubling time for total electricity generating capacity is projected by the Federal Power Commission [4] to be about twelve years in 2000 A.D. relative to about ten years in 1960, such an abrupt change in the doubling time after 2000 A.D. does not seem realistic. A modified electric power capacity projection that was recently presented by Swartout [2] in testimony before the Joint Committee on Atomic Energy shows a more rapid growth in the period before 1990 and a more gradual decrease in growth rate after 2000 A.D. The two curves are shown for comparison in Fig. 2.1 This new AEC projected growth curve has been used as the basis for studies presented in this paper.

Before leaving the discussion of the projected growth of nuclear power in the United States, some discussion of the factors that will affect the rate of growth is, perhaps, appropriate.

1. Nuclear power plants will replace coal stations only if the cost of nuclear power can be less than that of the coal-produced power in a particular region. The fraction of the country's power that will be produced by nuclear power will then depend on the fraction of coal-produced power that is higher in cost than that achievable with nuclear power. As will be shown later, approximately half of today's electric generating market could be provided by nuclear power if a fuel cycle

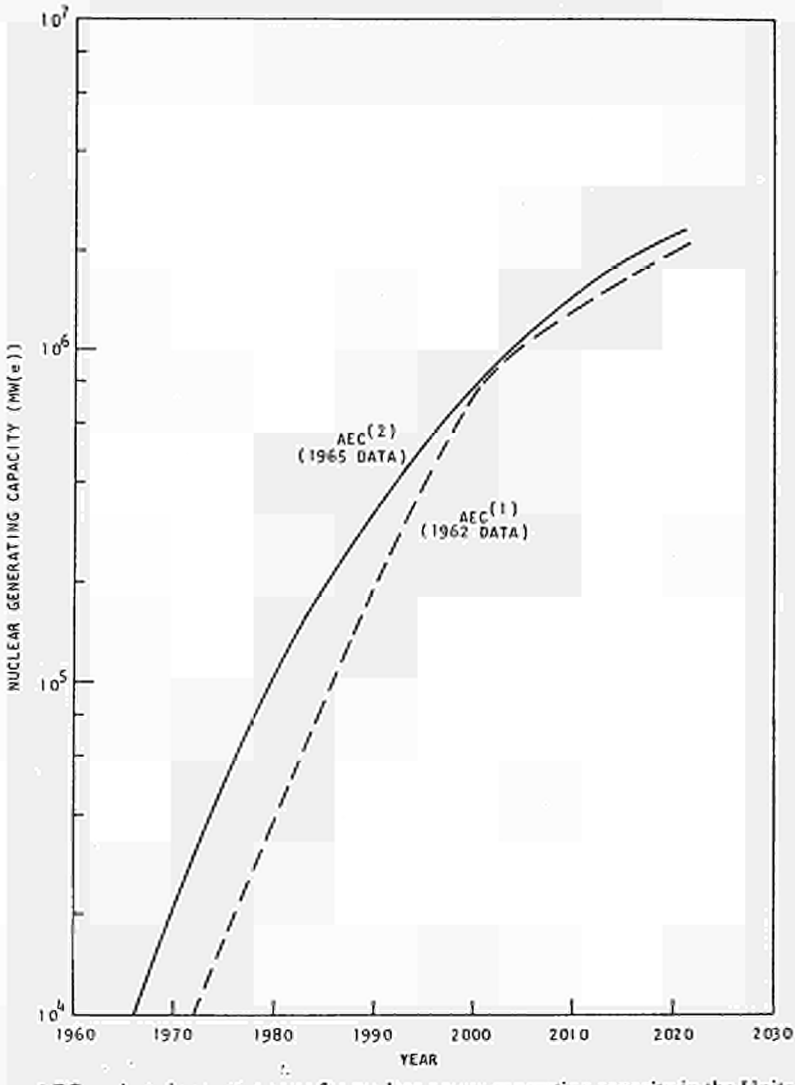

FIG. 2.1. — AEC projected growth curves for nuclear power generating capacity in the United States.

cost below 1.7 mills/kWh could be assured. This appears to be possible, even with the relatively inefficient converter reactors now being planned and built. Hence, one would expect, other factors allowing, that the nuclear power industry would grow rapidly to 50 % of the total generating capacity if a fuel cycle cost of 1.7 mills/kWh can be assured. It should grow still further if the fuel cycle cost decreases still more relative to coal costs.

2. The above arguments assume that coal costs will not decrease appreciably in at least the upper 50 % of the power cost range. Actually, with improved coal mining, transportation, burning, and electric transmission technology, the target

fuel cycle cost may decrease to less than 1.7 mills/kWh, so that nuclear power plants may have to do even better than indicated to gain wide acceptance.

3. The cost of electricity with nuclear power is most favorable when the size of the station is large. This could again impose a limitation on the growth of nuclear power, since much of the power generating capacity today is provided by relatively small power stations. However, with a power growth having a doubling time of about ten years, it can be seen that much larger power generation stations should be common within a few decades. Furthermore, the trend toward long-distance transmission and transmission line interconnections will tend to encourage the use of larger central station power plants.
4. The fuel cycle cost achievable with the nuclear power plants must remain below the target or "critical fuel cycle cost" independent of changes in uranium ore and production costs. This will be discussed in some detail in Sections 4 and 5 of this report.
5. The rate of nuclear power growth will also depend on the confidence of the utility industry in the economics, reliability, and safety of nuclear plants. Hence, there may be some time lag between the demonstration of a new reactor concept and its general acceptance by the utility industry.

With the exception of fuel cycle economics, these factors will not be discussed further in this report. We therefore depend on more careful analyses by other sources for the validity of the assumed nuclear power growth curves. One such analysis has recently been reported by the Federal Power Commission [4]. However, it is again emphasized that the fuel cycle cost for nuclear power plants is of great importance in assuring acceptance of nuclear power, and this subject will receive considerable attention in this report.

TABLE 2.1. — Uranium and thorium * resources in the United States (1)

Cost Range ($/lb of U_3O_8)	Reasonably Assured Resources (10^6 metric tons)		Estimated Total Resources (10^6 metric tons)	
	Uranium	Thorium	Uranium	Thorium
5-10	0.3	0.1 *	0.6	0.3 *
10-30	0.3	0.1 *	0.5	0.15 *
30-50	3.8	2.4	6.2	8.0
50-100	4.6	6.4	12	20
100-500	380	800	1,500	2,400
11-22 **			4,000	

* Incomplete estimates exist for thorium resources at recovery prices below $30 per lb.

** Estimated cost of recovery from sea water ([5]).

The basic data on recoverable uranium and thorium reserves, as outlined by the AEC report [1, 2] are used throughout this analysis. Since it is more convenient to have the reserves expressed in metric ton units of metal, the data summarized in Table 2.1 are expressed in these units. The last line in the table, however, refers to the estimated cost of separating uranium from sea water [5]. If this process proves to be feasible for the costs estimated, this development would have important implications on the necessity for developing fast breeder reactors. This will be discussed more completely in Section 4.

The important point to be gained from an examination of the resource data is that the total quantity of recoverable nuclear resources is enormously large. Consequently, to achieve the maximum utilization of our nuclear resources, the primary problem is to find a way to use the resources economically, in spite of the cost of recovery. Hence, conservation of resources, per se, is only of interest in so far as good conservation can delay the time when it will be necessary to use the more expensive ores. Even this consideration is of only minor long-range importance, as will be seen in the following section.

3. — Uranium requirements of various reactor concepts

In a growing nuclear power economy the fuel requirements depend both on :

1. The increase in fuel inventory required to start up new reactors, and
2. The fuel required to replace the net fuel consumed in generating energy.

The fuel requirements to allow for new reactor startups depend, of course, on the growth rate of the total nuclear capacity and on how much fuel inventory is held up by the reactors. The inventory requirement for a nuclear plant is inversely proportional to the system specific power measured by the kilowatts of electricity generated per kilogram of fuel held up both in the reactor and in the fuel fabrication and reprocessing plants. Specific power is more usually specified in units of kilowatts of heat per kilogram of fuel in the reactor core, i.e., kW(t)/kg. In comparing the inventory utilization of different reactor concepts, it is necessary, then to adjust the specific power in kW(t)/kg for the thermal efficiency of the plant and the fuel turnaround time relative to the irradiation time. For example, a reactor with an apparently high specific power of 2,000 kW/kg, a fuel life of two years, a fuel turnaround time of one year, and a thermodynamic efficiency of 30 % has an effective system specific power of 400 kW(e) kg of fuel held up. In contrast, a reactor with a more modest specific power of 1,250 kW/kg, but with a fuel life of four years and an efficiency of 45 % would have an effective system specific power of 450 kW(e)/kg of fuel held up. Hence, the higher efficiency and longer fuel life for the second reactor would more than make up for the higher apparent specific power of the first reactor. Furthermore, the low efficiency of the first plant would impose an additional penalty on the utilization of the fuel resources in the fuel burnup requirements.

The net fuel consumption depends on the conversion ratio of the reactor. A reactor system having a conversion ratio of 1.00 at equilibrium, including allowance for fuel losses in reprocessing, would be self-sustaining, i.e., would require no external fuel feed makeup after the reactor system had reached equilibrium. Obviously, a self-sustaining reactor is of only minor importance in a growing power economy. For a reactor system to be truly self-sustaining in a growing nuclear power economy, it would be necessary that the yearly breeding gain relative to the total inventory of the system be equal to the nuclear power growth rate. Hence, both the breeding ratio and specific power of the system must be considered simultaneously. The argument that a self-sustaining reactor is of interest for the long-range future when the power economy has reached a steady state would appear to be fallacious, since the growth rate is unlikely to decrease to zero for at least a century, if ever. Aside from resource considerations, reactors with low specific powers are of little interest, since they tend to be uneconomic.

Therefore, both system specific power and conversion ratio, or breeding ratio, are important considerations in choosing a reactor concept to meet the long-range objectives. The relative importance of these two characteristics in the conservation of nuclear fuel will be illustrated for several typical reactor conditions in the succeeding discussion. It is again emphasized, however, that conservation of the nuclear fuel resources in itself is not of over-riding concern in the goal of maximum utilization. Of greater importance, as will be amplified later, is the economic use of a substantial fraction of all the recoverable resources.

Figure 3.1 indicates the inventory and burnup requirements for uranium in a reactor system typical of today's pressurized and boiling water reactors (BWR). For this example, a system specific power of 800 kW/kg and a thermal efficiency of 32 % were assumed. The system specific power is based on the assumption that the fuel processing requires one year outside the reactor for each reactor cycle. Typical BWR plants with some stretch capability appear to have equilibrium loading conditions that lead to a reactor specific power of about 800 kW/kg and a system specific power of about 700 kW/kg. Projected pressurized water reactor (PWR) plants are expected to have an equilibrium feed reactor specific power of about 1,200 kW/kg, but a system specific power of about 900 kW/kg. The bottom curve in Fig. 3.1 represents the cumulative number of metric tons that would have to be mined simply to put new reactors into service to meet the growing demand for power under the assumptions indicated in the previous section.

In addition to the uranium requirements for starting up new plants, uranium is also required to replenish that consumed in generating the energy. For each gram of fuel consumed, CR grams of fertile material will be converted to new fuel. (CR is defined as the net conversion ratio for the reactor system.) Hence, the net fuel burnup is proportional to 1-CR. The electric energy produced per gram of fuel consumed is, of course, proportional to the thermal efficiency of the plant. The difference between the top curve in Fig. 3.1 and the inventory curve represents the amount of uranium that would have to be mined to meet the burnup requirements

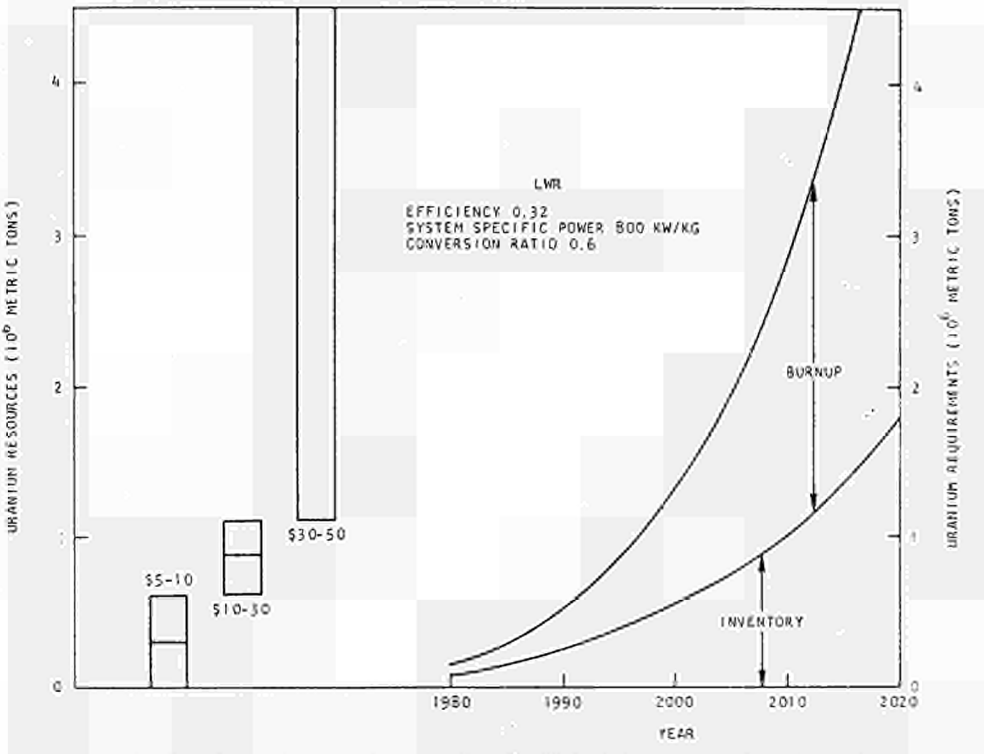

FIG. 3.1. — Projected uranium requirements for the United States assuming all nuclear power to be supplied by light water reactors.

for the reactor conditions specified. A conversion ratio of 0.6 was assumed for these calculations; this value is probably reasonable for projected light-water-moderated reactors using low-enrichment uranium.

It has been assumed in the calculation of the burnup requirements that all of the plutonium produced is recycled. If the plutonium is not recycled, the uranium requirements would be approximately twice that shown for fuel burnup, even after allowing for the plutonium that is burned before fuel discharge in the first cycle.

On the left side of Fig. 3.1, the amounts of uranium recoverable in different price ranges are shown, based on data from the AEC report [1]. The bottom segment of the bar represents the assured reserves and the top segment the total reserves estimated for each price range. It can be seen that a continuing trend of building the present-day inefficient reactors would exhaust the low-cost ores shortly after the year 1990. Indeed, reactors of this type that are constructed after 1970 would almost certainly have to be operated for some part of their plant lifetime on uranium ore costing more than \$10 per pound of U_3O_8. Hence, it can be seen that the near-term low cost of \$5-\$6 per pound for ore is not a good basis for evaluating the long-range economic potential of this type of reactor. It can be argued that more vigorous prospecting activities will probably uncover substantially more low-cost ore deposits.

However, because of the fast growth anticipated for nuclear power, doubling the amount of ore only postpones the day of reckoning by about five years. Even if the amount of low-cost uranium is an order of magnitude higher than that estimated, the low efficiency converters could survive for only a few more decades. While new discoveries could change the degree of the problem slightly, only a major breakthrough, such as the economic separation of uranium from sea water, could change the over-all picture significantly. However, it is not likely that this development could assure uranium resources at costs less than $10 per pound of ore. Clearly then, either we must look to other types of reactors to postpone the time when we will exhaust our low-cost uranium ore, or we must design our reactors to utilize the more expensive ores economically.

In this section, we will be primarily concerned with an examination of a few reactor types to see how effective these reactors might be in conserving the low-cost uranium ores. We will not give particular attention to combinations of reactors in this section, and will not question where recycle fuel might be obtained for the fast reactors that operate most effectively with plutonium. This will be discussed in another section. The economics of reactor operation with the more expensive ores will also be discussed in another section of this report. It will be seen from succeeding comparisons that the HTGR and high-performance fast breeder reactors are, indeed, more effective in conserving the uranium resources than are the light-water and heavy-water reactors using the low-enrichment-uranium cycle.

Figure 3.2 shows the inventory and burnup requirements assuming all nuclear power in the future is generated from the HTGR plants. First of all, it is interesting to note that the difference between systems having conversion ratios of 0.95 and 0.90 is not very significant, since the predominant requirement, with such excellent neutron economy, is for supplying the fuel inventory to start up new reactors in the expanding nuclear power industry. It also is noted that the better characteristics of the HTGR result in uranium requirements that are only about one-third of the requirements previously shown for the less efficient reactors. In spite of this improvement, however, the critical date for exhaustion of uranium at any particular ore cost is delayed only about ten years.

The HTGR resource requirements shown in Fig. 3.2 are based on a fuel element design using BeO spines. If an all-graphite fuel element is assumed, the optimum conversion ratio is about 0.85 instead of approximately 0.95. However, the optimum specific power for the all-graphite core tends to be somewhat higher, and the total uranium requirements are about the same. Some improvements in the HTGR uranium requirements might be realized by future design developments, such as the controlled release of volatile fission product poisons. However, it is not likely that the total uranium requirements would be changed substantially. As will be shown in the subsequent discussion, it is also unlikely that other reactor systems could improve significantly, if at all, over the HTGR unless very-high-performance fast-breeder reactors should become economically and technically feasible within the next twenty years.

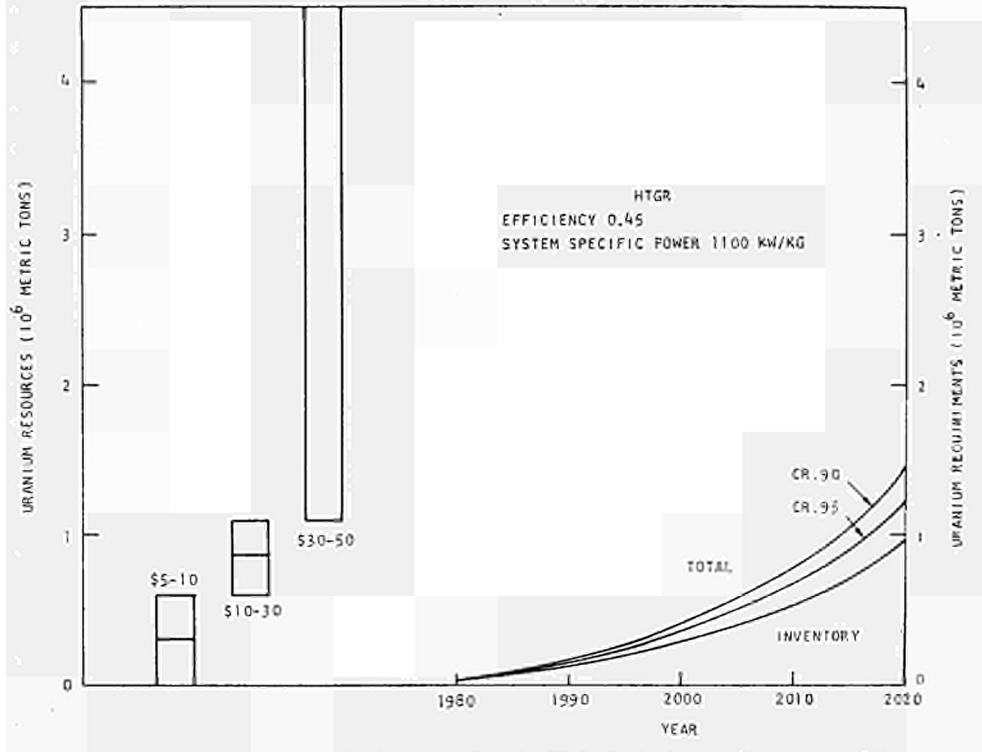

FIG. 3.2. — Projected uranium requirements for the United States assuming all nuclear power to be supplied by HTGR plants.

It is, perhaps, of special significance to examine the fuel requirements for a heavy-water reactor, since much attention [3, 6, 7] has been given to its potential for high specific power and good neutron economy. Several versions of the heavy-water reactor (HWR) have been developed or proposed involving different coolants and different fuel cycles. This discussion will be limited primarily to the heavy-water-cooled, heavy-water reactor described by the DuPont Laboratory [6, 8] and reviewed in the *Comparative Evaluation of Advanced Converters* by the Oak Ridge staff [9] .Based on reported data, the following characteristics were used for the HWR :

Reactor specific power, kW/kg	4000
System specific power, kW/kg	.2000
Thermal efficiency	0.267
Conversion ratio	0.7

Fig. 3.3 illustrates the uranium requirements for a power economy consisting entirely of HWR plants. It can be seen from the figure that the inventory requirements of a heavy-water reactor tend to be very modest, whereas most of the uranium requirements arise from the net fuel burnup. Some consideration [8] has also been given to the U^{233}/Th^{232} cycle in the heavy water reactor. Although the net fuel burnup is considerably better for this fuel cycle, the inventory requirements are

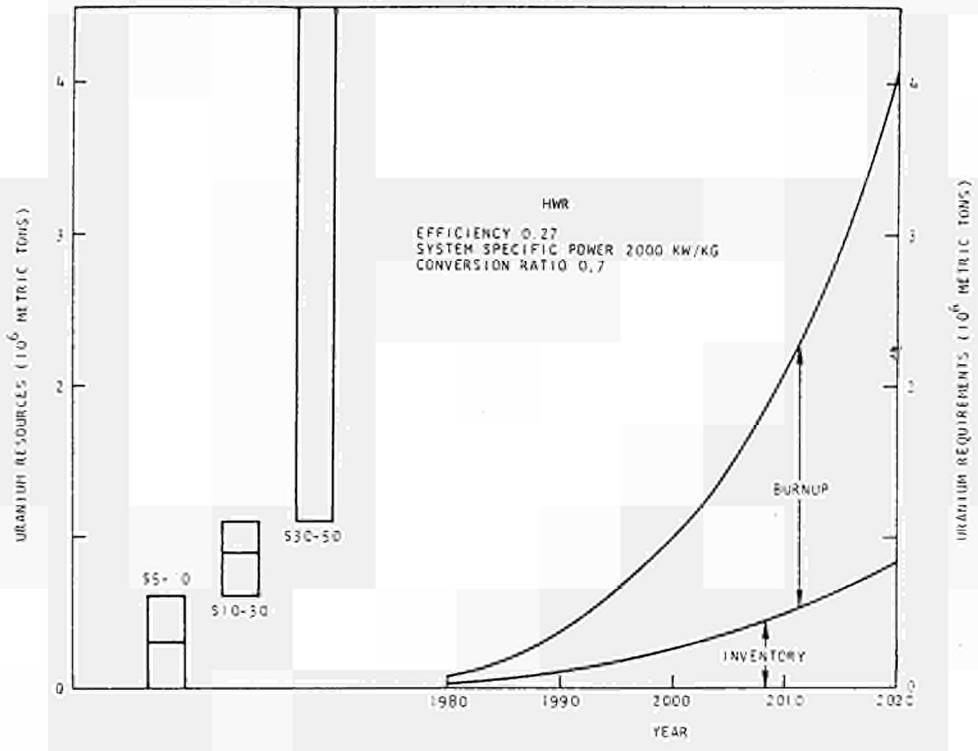

FIG. 3.3. — Projected uranium requirements for the United States assuming all nuclear power to be supplied by heavy water reactors.

somewhat larger because of the low thermal efficiency and the limitation on specific power that is imposed by neutron losses to Pa^{233}. Furthermore, the fuel cycle costs tend to be higher for the U^{233}/Th^{232} cycle, so that there is little incentive to use this cycle in the heavy-water reactor.

The organic-cooled, heavy-water reactor (OHWR) [7] shows approximately the same over-all uranium requirements as the heavy-water-cooled, heavy-water reactor. Whereas the thermal efficiency of the OHWR is higher than the HWR, the specific power is lower, so that the effects compensate. In general, it can be seen that the uranium requirements for the heavy-water reactors are somewhat lower than those for the light-water reactors, but are significantly higher than for the HTGR.

Before leaving the subject of uranium conservation, it is of some interest to examine the performance of the fast-breeder reactors (FBR). This evaluation is even more difficult because of the very large uncertainties in the operating characteristics of this type of reactor arising particularly from materials problems and safety considerations. Because of heat transfer and physics considerations, it is generally more difficult to achieve a large specific power in the fast-spectrum reactor than in the thermal-spectrum reactor. Furthermore, it may be necessary to degrade the spectrum in at least some types of fast breeder reactors, in order to enhance the

Doppler coefficient sufficiently to assure safe operating characteristics. Under these conditions, the breeding gain may optimistically be about 1.3, and possibly even smaller. A burnup time of 100,000 MWd/T has generally been established as an objective for the fast breeder reactors using uranium and plutonium oxide fuel elements, but it is possible that materials damage problems could limit burnup times to less than 50,000 MWd/T, thereby making the out-of-reactor inventory about as large as that in the reactor.

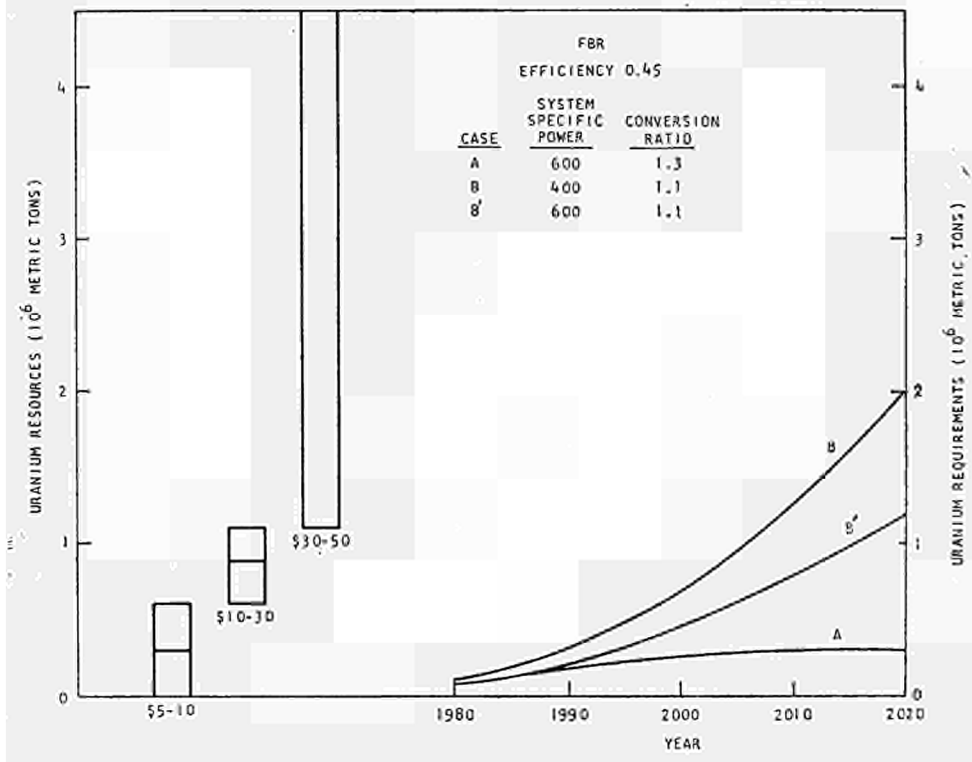

FIG. 3.4. — Projected uranium requirements for the United States assuming all nuclear power to be supplied by fast breeder reactors.

Fig. 3.4 shows the uranium requirements that might be expected of fast-breeder reactors under various assumed conditions, if plutonium resources were immediately convertible from the U^{235} in uranium ore to start up the fast reactors. Curve A in the figure indicates the uranium requirements assuming the FBR plants have, on the average, a conversion ratio of 1.3, a reactor specific power of 800 kW/kg, and a fuel exposure time of about 100,000 MWd/T. Curve B shows the requirements should it be necessary to degrade the conversion ratio to 1.1 and the fuel exposure time to 50,000 MWd/T to avoid safety and materials damage problems. Curve B' indicates the effect of degrading only the conversion ratio.

Under the assumption of immediate availability of plutonium from U^{235} with no loss of fissile material, it would appear from this analysis that the FBR typified by conditions A could utilize the resources quite effectively. In Section 5 we will examine the uranium requirements of a reactor system consisting of a converter and a breeder reactor. Under these more practical conditions, it will be seen that the total uranium requirements of the FBR will be affected quite strongly by the type of converter reactor used to supply the plutonium for the FBR. Under these more practical conditions, it is very probable that the low-cost uranium ores would be exhausted even with breeder reactors. The more important subject is, then, the potential economic performance of the various reactor concepts under varying conditions of uranium ore cost.

Consequently, the next section will deal with the economics of the various reactor systems under different assumptions of the nuclear ore cost. In the final analysis, our economic system will naturally select the reactor concept or concepts that will offer the most advantageous economics. If this system can maintain its economic advantage in the face of rising uranium ore costs, then, and only then, will our nuclear resources be utilized to the maximum extent.

4. — Fuel cycle economics of various reactor concepts as affected by changing uranium ore costs

Nuclear power can contribute to the rapidly growing energy economy if the cost of generating nuclear power becomes and remains competitive with the cost of power from other sources, primarily the fossil fuels. While the discussion in this report is limited to the component of power cost associated with the fuel cycle, the fuel cycle cost objectives can be put in proper perspective by some reference also to the relative capital costs of fossil-fired and nuclear power plants. On the basis of experience to date, it appears that the capital cost of a large fossil-fired power station will be approximately \$20/kW cheaper than that of a similar nuclear power station for a number of years. Consequently, in a private power economy the fuel cycle cost of a large nuclear power station must be at least 0.4 mill/kW-hr lower than that of a plant using fossil fuels. Approximately 50 % of the installed generating capacity in the United States uses coal having an energy cost of $23¢/10^6$ Btu, or greater. When used in a typical modern plant with a thermal efficiency of about 38 %, coal at $23¢/10^6$ Btu will generate electric power having a fuel cost component of about 2.1 mills/kW-hr. Nuclear power plants should, therefore, promise a fuel cycle cost of no more than 1.7 mills/kW-hr to be economically competitive with about 50 % of our coal-fired power plants. Throughout our discussion of nuclear power economics we will, then, refer to 1.7 mills/kW-hr as a critical number for the acceptance of nuclear power stations. Obviously, reactors having higher capital costs would have to show an even lower fuel cycle cost.

It will be seen in the subsequent discussion that large nuclear power plants should have no difficulty in achieving fuel cycle costs below 1.7 mills/kW-hr, at least

for the next two decades. This objective should, in fact, be made easier by the near-term decreases in uranium ore cost that are expected. In the long range, however, the uranium ore cost is expected to increase as the higher grade ores are exhausted. Under these conditions the critical fuel cycle cost becomes increasingly hard to meet, and the reactor performance characteristics become increasingly important.

In this section the importance of the various reactor performance characteristics will be examined by observing their effects on the fuel cycle cost for some very elementary reactor examples. The effect of ore costs on fuel cycle costs will then be illustrated for a few typical reactor conditions, and finally, the effects of probable economic and fuel availability trends will be examined as a basis for selecting appropriate combinations of reactor concepts to utilize uranium economically over the long range.

The relative importance of thermal efficiency, specific power, conversion ratio, and fuel burnup time on fuel cycle economics can be illustrated quite graphically by observing the cost effect of degrading successively each of these reactor performance characteristics in a simple recycle reactor. The following arbitrary economic assumptions have been made for these simplified fuel cycle calculations :

1. The fabrication charge is \$100 per kg of metal.
2. The reprocessing and shipping charges are \$50 per kg of metal.
3. The Th/U ratio is 30.
4. The interest rate is 10 %.
5. The fuel turnaround time is 1 year.
6. The fuel value is \$14 per gram of fissile material.

With these assumptions and the assumed reactor performance characteristics shown at the top of Table 4.1, the fuel cycle cost components and total fuel cycle costs for six sample cases are shown in the bottom part of the table. Case A represents a reactor having a reasonably good performance in all four areas. Case B shows the effect of degrading the thermal efficiency, Case C the specific power, Case D the conversion ratio, and Case E the fuel burnup time. Case F shows the effect of degrading two characteristics simultaneously, in this case the thermal efficiency and the fuel lifetime. As can be seen, the fuel cycle cost penalty is between 0.2 and 0.5 mill/kW-hr for each of the single degradations, and almost 1.2 mills/kW-hr for the double degradation. Clearly, it is desirable to design a reactor with good performance characteristics in all four areas simultaneously in order to achieve the best possible economic performance. In this respect, the HTGR excels as an advanced converter.

It was pointed out in the previous section that the requirements for uranium ore will not exceed the availability of low-cost deposits for about 30 years, even if relatively inefficient converters are used for nuclear power plants. In fact, because of the surplus of uranium ore that now exists, cheaper mining opreations, and the introduction of toll enrichment, the cost of uranium ore is expected to fall from \$8 per pound to \$5 to \$6 per pound almost immediately and remain there for 15-20 years.

TABLE 4.1. — Effect of reactor performance characteristics on fuel cycle cost (U^{233}/Th^{232} Recycle Reactors)

	Case A	Case B	Case C	Case D	Case E	Case F
Thermal efficiency, %	45	30	45	45	45	30
Initial specific power, kW/kg	1,000	1,000	500	1,000	1,000	1,000
Conversion ratio	1.00	1.00	1.00	0.80	1.00	1.00
Fuel life, years	4	4	8	4	2	2
Fabrication	0.25	0.37	0.25	0.25	0.50	0.74
Reprocessing	0.12	0.18	0.12	0.12	0.25	0.37
Depletion	0	0	0	0.31	0	0
Working capital	0.56	0.83	1.00	0.48	0.67	1.00
TOTAL, mills/kW-hr	0.93	1.38	1.37	1.16	1.42	2.11

Taking a more long-range point of view, it would be expected that the cost of uranium ore would rise quite rapidly after 1995 as the low-cost, high-grade uranium ore deposits are exhausted. Although this may seem rather far in the future, it must be recalled that reactors built after about 1980 will probably have to use the higher cost uranium for some significant fraction of their normal operating lifetime. Consequently, it is pertinent to examine the fuel cycle cost behavior of reactor concepts now being developed under different assumptions on the uranium ore cost. In this context we will look at the effect of uranium ore cost on fuel cycle costs for a light water reactor, a heavy water reactor, the HTGR, and some fast breeder reactors.

The economic assumptions and reactor characteristics used for the comparison of the thermal reactors are summarized in Table 4.2. The data are generally consistent with information contained in the Oak Ridge evaluation report [9] for advanced converter reactors and with other published reactor data, although some of the data have been simplified and rounded off. The fuel fabrication costs and reprocessing costs have been deliberately chosen to be very low, reflecting the state of technology that could possibly exist in another 15 to 25 years.

Fig. 4.1 shows the fuel cycle costs for the LWR, HWR, and HTGR for different uranium ore cost assumptions. The first two reactors are both low-enrichment-uranium reactors while the HTGR is assumed to use the Th/U^{233} cycle with U^{235} makeup. The D_2O working capital and makeup charges have been added to the fuel cycle cost of the heavy water reactor. It can be seen that the projected fuel cycle cost for the HTGR is uniformly lower than that of the low-enrichment reactors. Furthermore, the increase in cost per unit rise in ore cost is considerably smaller for the HTGR than for the low-enrichment reactors, It is noted that the fuel cycle costs for the low-enrichment reactors all cross the critical cost line in the ore cost

TABLE 4.2. — Economic assumptions and reactor performance data used in economic evaluations

Uranium ore cost	Variable
Separative cost, $/kg	30
U^{233}/U^{235} value ratio	14/12
$Pu^{239} + Pu^{241}/U^{235}$ value ratio	10/12
D_2O cost, $/kg	44
Finished graphite cost, $/kg	6
Working capital interest rate, %	10
Fuel turnaround time, years	1

	LWR	HWR	HTGR
Fuel cycle	U/Pu	U/Pu	Th/U
Fabrication cost, $/kg	50	20	100
Shipping and reprocessing cost, $/kg	30	20	50
Fuel burnup, MWd/kg	22	15	~60
Fuel burnup, years	~4	~1	4
Initial specific power, kW/kg	~700	~4,000	~1,300
Conversion ratio	~0.6	~0.7	~0.85
Thermal efficiency	0.32	0.267	0.45

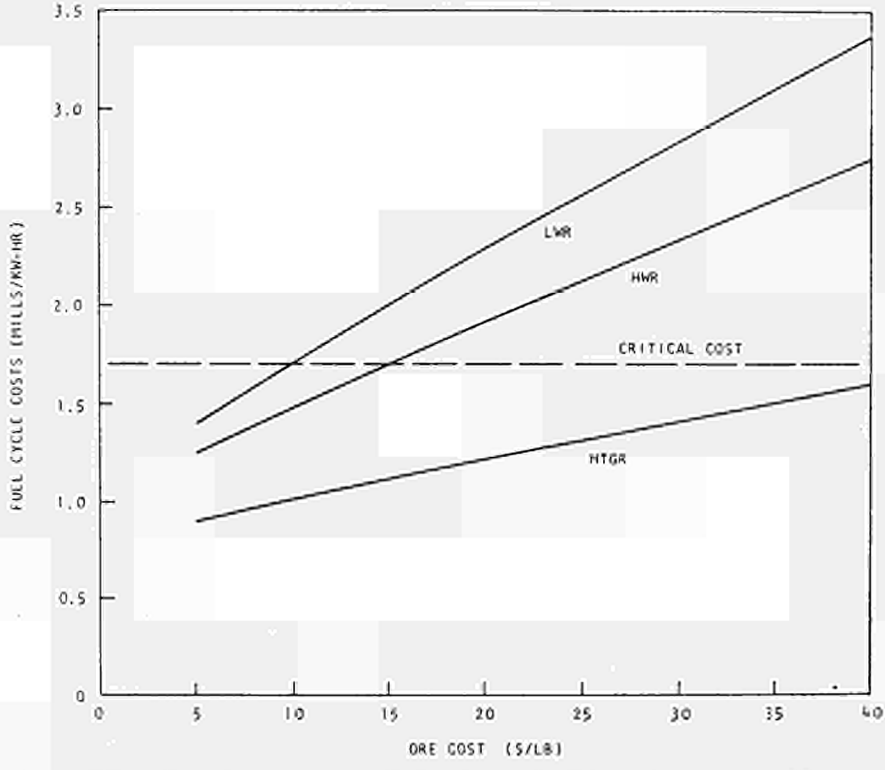

FIG. 4.1. — Effect of uranium ore cost on fuel cycle cost for light water reactor and heavy water reactor using low enrichment uranium and the HTGR using the Th/U^{233} recycle.

range of \$10 to \$15 per pound. The HTGR costs appear to be competitive, relative to the critical cost, for uranium ore costs well in excess of \$30 per pound. A recent report by Davies, *et al.* [5], states that laboratory experiments have shown that the enormous uranium resources in sea water can apparently be recovered at costs in the neighbourhood of about \$20 per pound. The HTGR fuel cycle cost using \$20 per pound uranium ore is seen to be sufficiently attractive to suggest that the HTGR can be a long-range solution to the energy production problem.

It has frequently been suggested that the HWR (and possibly the LWR) might benefit in the long range by using the Th/U^{233} fuel cycle instead of the low-enrichment-uranium fuel cycle. Fig. 4.2 shows the fuel cycle costs (again including D_2O charges) as a function of ore cost for the HWR low-enrichment reactor and the HWR Th/U^{233} recycle reactor. The HTGR is again shown for comparison. It can be seen that the Th/U^{233} cycle for the HWR is less attractive than the low-enrichment-uranium cycle for uranium ore costs up to about \$20 per pound. At this point the fuel cycle cost has exceeded the critical cost, so that it is doubtful that recycle operations

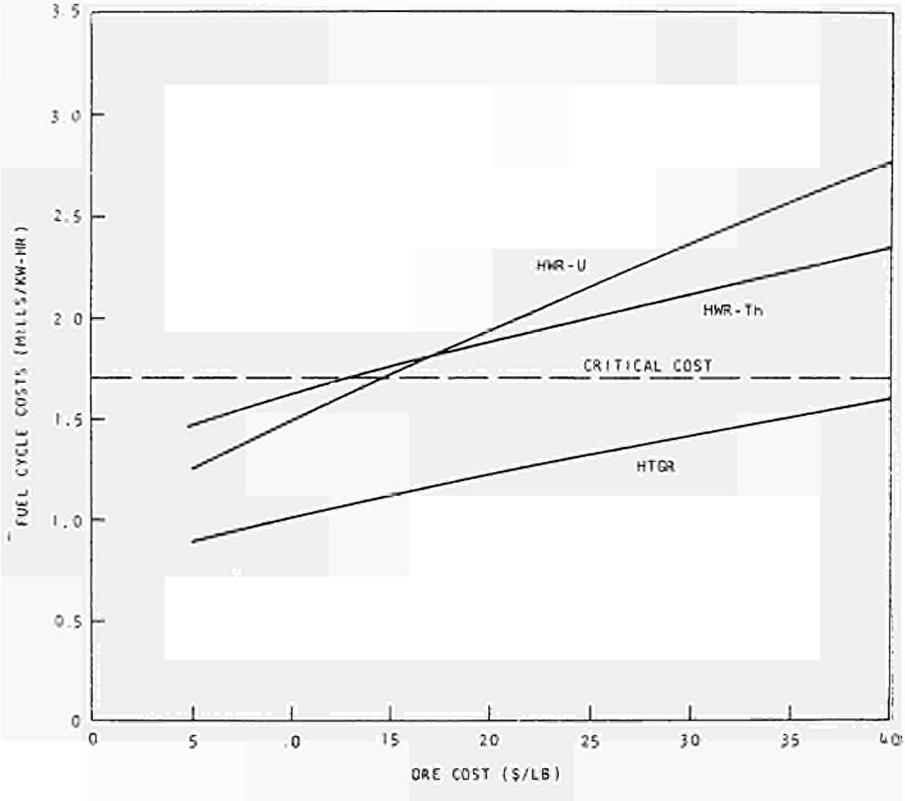

FIG. 4.2. — Effect of uranium ore cost on fuel cycle cost for heavy water reactors using low enrichment uranium and Th/U^{233} recycle and the HTGR using the Th/U^{233} recycle.

will ever be attractive in the HWR. While results are not shown for the LWR, the same general behavior is found for this reactor also.

Although the HTGR appears to be capable of solving the long-range energy supply problem, the HTGR will become a long-range solution only if its power cost promises to be lower than that of other power plants. The strongest competitor is undoubtedly the fast breeder reactor. Hence, some attention has also been given to the fuel cycle costs of various fast breeder reactors relative to the HTGR. An accurate appraisal of the fast breeder reactor is extremely difficult because of the many uncertainties in materials, physics, and safety problems. Some attempt has been made to estimate the effect of uranium ore cost on fuel cycle cost for the fast breeder reactors by using some of the results from the recent fast breeder reactor studies conducted by U. S. contractors [10] for the AEC as a base point. Hence, typical fabrication and reprocessing charges reported for oxide fuel elements have been used in these evaluations, and a conversion ratio of 1.30, a specific power of of 800 kW/kg, and a fuel exposure of 100,000 MWd/T have been assumed. These estimates appear to be optimistic objectives for the fast-spectrum, sodium-cooled reactor. Assuming that the value of fissionable plutonium is 10/12 that of U^{235} (possibly an overly optimistic low value if there is a strong demand for Pu) and assuming

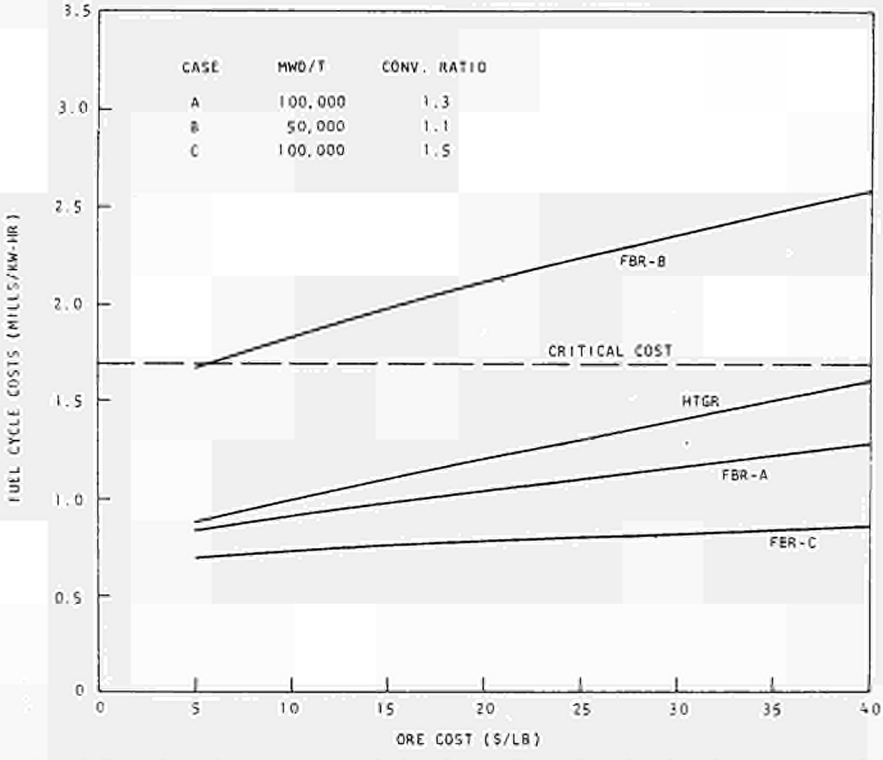

FIG. 4.3. — Effect of uranium ore cost on fuel cycle cost for various fast breeder reactor conditions relative to fuel cycle cost for HTGR.

private financing, the fuel cycle cost as a function of uranium ore cost is shown by Curve A in Fig. 4.3. Under these favorable conditions, the fuel cycle cost is about 0.1 mill/kW-hr lower than that of the HTGR for ore at $6 per pound and about 0.2 mill/kW-hr lower for ore at $20 per pound. Hence, under these favorable conditions, the fast breeder reactor would be competitive with the HTGR if the capital cost of the FBR does not exceed that of the HTGR by more than about $10/kW(e).

Curve B in Fig. 4.3 indicates the fuel cycle cost for the same reactor if the conversion ratio and fuel exposure time must be degraded in order to satisfy safety and materials problems. In this case, it is seen that the fuel cycle cost for the fast breeder reactor is significantly poorer than that for the HTGR.

Curve C indicates the fuel cycle cost data for the case where the conversion ratio is 1.5 and the other performance characteristics are the same as assumed for Case A. These charateristics are typical of the objectives for the fast gas-cooled reactor using oxide-type fuel elements. This case clearly has the potential for showing a significant improvement over the HTGR in the long range and would seem to warrant a continuing development effort.

In the fuel cycle cost data presented for the HTGR, we have, until now, shown only the results of one set of core design conditions and economic assumptions. The detailed design specifications for an HTGR core would, of course, depend on many technical and economic considerations. A very large number of survey calculations have been done on the 1,000-MW(e) HTGR to define the range of interest for the various design parameters. Some of the results of these studies will be illustrated.

The characteristics of the 1,000-MW(e) reactor are summarized in Table 4.3. Among the reactor characteristics that can be classified as independent variables are the following :

1. *Fuel Cycle.* The $U^{235}/Th^{232}/U^{233}$ cycle and the $U^{233}/Th^{232}/U^{233}$ recycle are of greatest interest for the HTGR. Studies have been made for the low-enrichment fuel cycle and for a recycle operation using Pu as makeup fuel. These cycles are the subject of a different paper.
2. *Fuel Element Design.* Two fuel element designs have been considered in the large HTGR studies. In one case, the fuel element incorporates a BeO spine whereas in the second, the entire moderator consists of graphite.
3. *Initial Fuel Loading.* The initial fuel loading is generally characterized by the ratio of moderator to fertile atoms, i.e., Be/Th or C/Th atom ratios.
4. *Fuel Residence Time.* Fuel residence times from three to ten years have been examined. In all cases it has been assumed that the reactor is refueled semiannually, with a fraction $1/2\ \tau$ of the fuel elements being replaced, where τ is the residence time.

For the studies reported here, the reactor power density has been chosen at 7 w/cm^3 and the core reflector has been chosen to be 61 cm thick. Some consideration has also been given to the use of thorium blankets at the edge of the core, and to the possible use of fuel elements designed to purge the volatile fission products. The

TABLE 4.3. — Summary of the physical characteristics of the target core

Power, MW(t)	2,340
Power, MW(e)	1,050
Coolant	Helium
Coolant inlet temperature, °F	720
Coolant outlet temperature, °F	1,470
Fuel element diameter, in	4.65
Fuel element pitch, in	4.7
Fuel element array	Triangular
Fuel element length, ft	20
Core diameter, ft	31.1
Core length, ft	15.5
Number of fuel elements per core	5,489

use of thorium blankets would enhance the conversion ratio by about 0.02 over the values reported in this paper.

An HTGR using fuel elements with BeO spines and a thorium blanket would be able to achieve a conversion ratio greater than unity. This mode of operation might be economically attractive when the cost of uranium ore becomes sufficiently large or under circumstances where it might be desirable to be independent of an enriched uranium supply. The use of fuel elements designed to remove the volatile fission products might be justified when more experience is available on the control of fission products.

Table 4.4 illustrates some typical conversion ratios calculated under various HTGR operating conditions. Thus, for a 1,000-MW(e) HTGR with an all-graphite moderator, a conversion ratio of 0.75 is calculated for the case where U^{235} is used as the initial fuel charge. If the discharged U^{233} is stored for the first 6 years and subsequent cores are loaded with the first generation U^{233}, a conversion ratio of 0.99 could be achieved in the second core using an all-graphite-moderated core and a thorium blanket. With fuel elements containing BeO spines, the conversion ratio could be as high as 1.05 under similar circumstances.

Normally, it has been the custom at General Atomic to calculate the neutron balances for equilibrium conditions, i.e., after the fuel has been recycled through the reactor a very large number of times and the heavy element isotopes have reached an equilibrium atomic distribution. The buildup of some of the undesirable heavy element isotopes can be controlled by a fuel management program that either recycles the bred U^{233} only one cycle with the feed fuel (the once-through recycle), or keeps the makeup fuel distinct from other fuel and uses it for only one cycle, but continuously recycles the bred fuel (the bred-fuel recycle). The third column in Table 4.4 shows the neutron balance for an equilibrium cycle using the once-through-recycle fuel management program. The fourth column shows a similar cycle, but

TABLE 4.4 — Neutron balances for various HTGR conditions

Initial fuel	U^{235}	U^{233} (1st cycle)	U^{233} Recycle - (Equilibrium) (U^{235} makeup)		
Moderator	C	C	C	C/BeO	C/BeO
Fuel lifetime, years	4	3	4	4	4
Volatile fission product control	← Retained →				Withdrawn
η	2.06	2.22	2.16	2.17	2.22
$\varepsilon\eta$	2.06	2.22	2.16	2.24	2.29
Losses :					
Leakage	0.05	0.02	0.05	0.04	0.04
Moderator	0.03	0.03	0.03	0.04	0.06
Pa^{233} ($\times 2$)	0.03	0.03	0.03	0.03	0.04
$U^{236} + Np^{237}$	0.02	—	0.01	—	—
Xe^{135}	0.04	0.04	0.04	0.04	—
Other F.P.P.	0.11	0.08	0.10	0.08	0.07
Control	0.03	0.03	0.03	0.02	0.02
TOTAL losses, L	0.31	0.23	0.29	0.25	0.23
$\varepsilon\eta$-1-L	0.75	0.99	0.87	0.99	1.06

with fuel elements containing BeO spines. With a thorium blanket, a conversion ratio of about 1.01 could be achieved for this case even for the equilibrium condition. The final column indicates the conversion ratio that might be achieved if the volatile fission products are withdrawn from the fuel elements.

The C/Th and Be/Th atom ratios are very important parameters. It is generally found that the optimum C/Th or Be/Th ratios depend on other factors, such as the cost of fuel, the cost of fuel fabrication and reprocessing, and temperature limitations on the fuel elements. We will not attempt to present a detailed description of the effect of this particular variable on reactor performance characteristics. Hence, in the following discussion, the C/Th and Be/Th ratios will be chosen to be typical values pertinent to a particular set of cost assumptions.

The optimum fuel exposure lifetime in the HTGR depends on the cost of the fuel fabrication and reprocessing and, to some extent, on the cost of the uranium fuel. For the next decade, for example, while fuel manufacturing technology is still being improved and while the volume of production is expected to be relatively small, the cost of fuel fabrication will be sufficiently high to encourage a relatively long fuel lifetime in the reactor. Since a shorter fuel lifetime results in a better conversion ratio and therefore a lower depletion cost component, it is economically beneficial to decrease the fuel lifetime, within limits, when the fabrication and reprocessing costs justify such a decrease.

Since it is too early to estimate the future cost trends accurately for fuel processing, we have prepared fuel cycle costs for several possible processing cost patterns. The assumptions are as follows :

Fabrication Cost ($/kg)	Shipping and Reprocessing Costs ($/kg)
300	150
200	100
100	50

In each case, an additional $500 per fuel element is assumed for the cost of the finished graphite pieces. There is no particular basis for the specific choice of the numbers, except that the range is expected to cover future costs, and the fabrication cost should certainly decrease with time and experience. The lowest cost shown on the figure corresponds approximately to the fabrication cost estimated by the Oak Ridge analysis [9].

Fig. 4.4 illustrates the fuel cycle costs as a function of fuel life-time for the above three different assumptions on fabrication and reprocessing costs. The data assume recycle operations with U^{235} makeup and with fuel elements containing only graphite as a moderator material. It can be seen that the optimum fuel lifetime is 4 to 5 years if the fabrication cost becomes $100 per kg, or less. However, with the higher fabrication cost, the optimum fuel lifetime is larger.

Fig. 4.5 shows the calculated conversion ratio as a function of fuel burnup time. In this figure results are shown both for the graphite fuel element and the graphite/BeO fuel element. As would be expected, the conversion ratio improves significantly as the fuel exposure time (and consequently the fission product inventory) is reduced. Remembering that the conversion ratio is improved by about 0.02 when a thorium blanket is included, it can be seen that a conversion ratio greater than unity can be achieved with the graphite/BeO fuel element and a fuel life of 3 years.

5. — Resource utilization in reactor combinations

The importance of the reactor performance characteristics on uranium conservation and uranium utilization has been examined in Sections 3 and 4. It was tacitly assumed in the previous sections that U^{233} would be available for starting up thermal spectrum advanced converter reactors and Pu would likewise be available for starting up fast breeder reactors in whatever quantity was required. In practice, both of these fuels must, of course, come from the discharge of previous reactor cycles, since neither exists in nature. The rate of introduction of the recycle reactors is, then, limited by the production rate of U^{233} or Pu in non-recycle reactors. A complete analysis of the utilization of nuclear resources must then include an evaluation of reactor systems involving some reactors that produce the desired fuels (i.e.

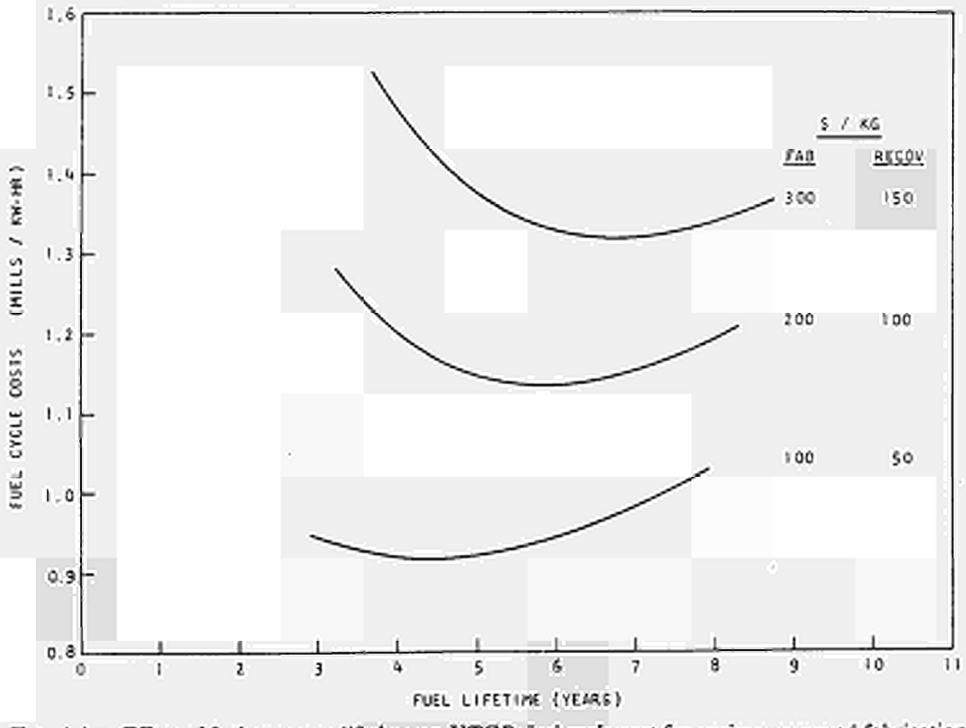

FIG. 4.4 — Effect of fuel exposure lifetime on HTGR fuel cycle cost for various assumed fabrication and recovery costs.

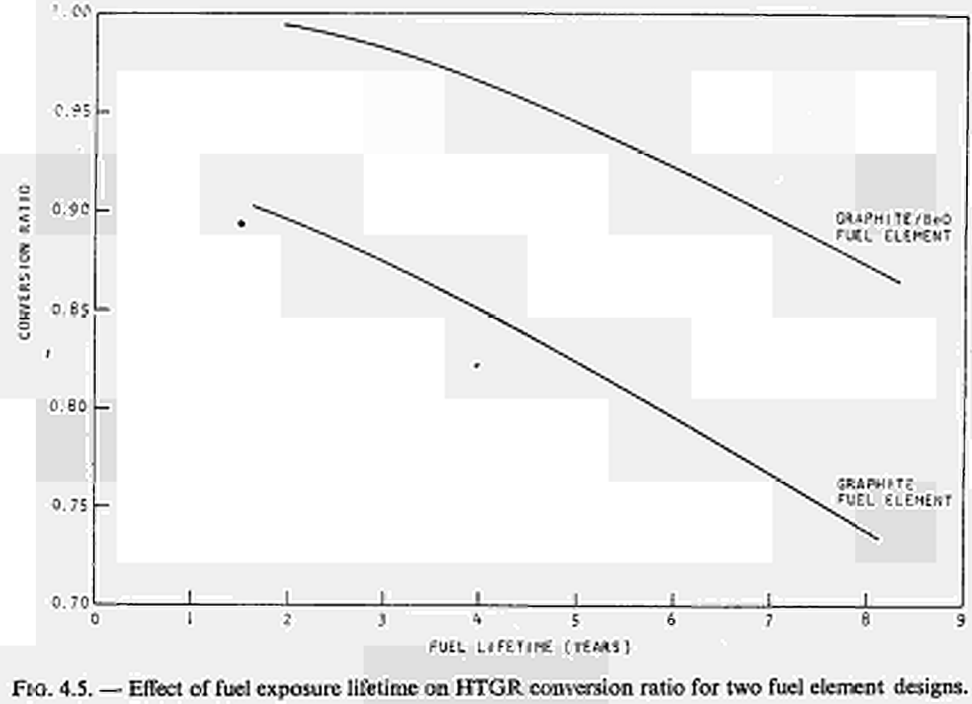

FIG. 4.5. — Effect of fuel exposure lifetime on HTGR conversion ratio for two fuel element designs.

feeder reactors) and reactors that use the bred fuels (i.e., fed or recycle reactors). A complete evaluation of these symbiotic systems represents a rather complicated operations research study, since it depends on detailed information of future nuclear fuel values, fuel supply and demand, and reactor operating characteristics and economics. Such an analysis is probably impractical at this time because of the limited amount of reliable information available. However, some general conclusions regarding the practicality of various symbiotic reactor combinations can be seen by some rather simple analyses of the uranium consumption and the fuel cycle economics for these combinations. The results of some of these studies will be presented in this section.

The startup of new reactor plants and the approach to recycle equilibrium of the new plants can be accomplished in several ways, two of which are :

1. Each reactor is initially fueled with U^{235} and either U^{238} or Th^{232}. The bred fuel is stored, kept separate from the fed fuel, and subsequently reused in the same reactor, with U^{235} makeup if necessary.
2. A symbiotic reactor system can be assumed in which some of the reactors are always fed with U^{235} and either U^{233} or Th^{232}. The bred fuel from these converter or feeder reactors is then used only in the fed or recycle reactors. New fed reactors can be started up only when fuel is available either from their own excess production or from the non-recycle feeder reactors. If more capacity is required than the fed reactors can provide, additional feeder reactors must be installed.

In this analysis, the second approach has been used, since it allows greater flexibility in optimizing the feeder-fed reactor system and indicates how current reactors can be used to produce the desired fuels. The growth curves for the installed capacity of nuclear power and the cumulative energy generated were covered in detail in Section II.

Two symbiotic systems involving HTGR feeders and HTGR recycle reactors were studied. The first of these, which resulted in a particularly small demand on nuclear resources, was a system consisting of

1. HTGR feeder plants using U^{235}/Th non-recycle, a fuel element design that incorporates BeO in the spines, a Be/Th atom ratio of 28, and a fuel residence time of three years.
2. HTGR fed plants using the U^{233}/Th recycle, fuel elements with BeO spines, a Be/Th ratio of 40, and a fuel residence time of three years.

The ratio of recycle to feeder reactors and the net resource requirements for this complex are shown in Fig. 5.1. When the capacity is growing very rapidly, about 60 % of it can be accommodated with recycle reactors. However, when the doubling time stretches out to ten years or more after year 2,000, the recycle reactors account for about 80 % of the capacity. The net uranium resource requirement by the year 2,020 is 1.1×10^6 metric tons, which is only slightly above that estimated to be available at \$5 to \$10 per pound.

A second HTGR system was considered that exhibits a somewhat larger nuclear resource commitment but probably operates with lower fuel cycle costs. This system is composed of

1. HTGR feeder plants using U^{235}/Th non-recycle, a fuel element design that uses only graphite as moderator, a C/Th ratio of 200, and a fuel residence time of four years.
2. HTGR fed plants using U^{233}/Th recycle, all-graphite fuel elements, a C/Th ratio of 200, and a fuel residence time of four years.

The ratio of recycle to feeder reactors and the net resource requirements for this system are also shown in Fig. 5.1. During the period of rapid growth, the recycle reactors can accommodate only about 30 % of the required capacity. When the growth rate slows down, roughly 50 % of the capacity can be accommodated by the recycle reactors. By the year 2,020, this reactor system requires roughly 1.9×10^6 metric tons of uranium resources. As a point of comparison, it is recalled that a light water reactor would require in excess of 5×10^6 metric tons of uranium.

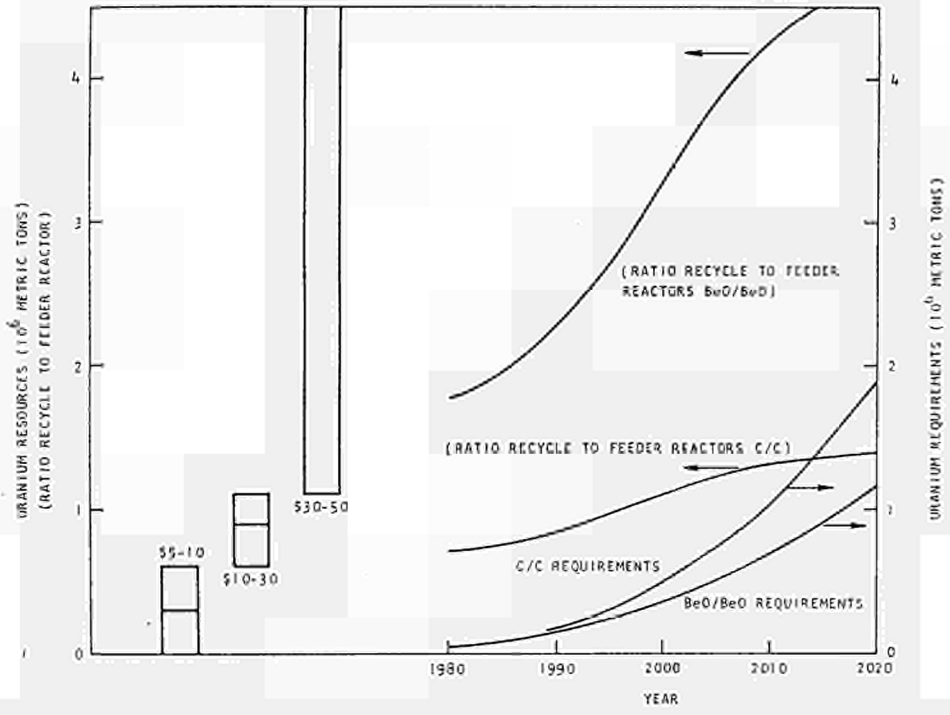

FIG. 5.1. — Uranium requirements and relative number of recycle to feeder reactors for two HTGR symbiotic systems.

The fuel cycle characteristics of these symbiotic reactors are shown in Table 5.1. In the case of the system using reactors with fuel elements having BeO spines, about 80 % of the energy up to the year 2,020, is generated in the very high conversion ratio, recycle reactors. Therefore, the average conversion ratio for the system is about 0.98, which is quite close to the equilibrium cycle value of 0.97 with U^{235} feed. For the system with all-graphite fuel elements, about 50 % of the energy up to the year

TABLE 5.1. — Fuel Cycle characteristics of symbiotic, 1,000-MW(e) HTGR complexes

C/Th (Be/Th)	(28) → (40)		200 → 200	
Fuel residence time, years	3	3	4	4
Fuel management	Non-recycle	Once-through	Non-recycle	Once-through
Conversion ratio	0.85	1.01	0.73	0.90
Eta	2.00	2.22	2.08	2.22
Burnup, fifa	0.7	1.6	1.4	2.0
Initial specific power, kW/kg	700	1,700	1,100	1,600
Initial fissile loading, kg	3,300	1,400	2,100	1,460
Net fissile requirements, kg/yr	630	0	480	90
Fissile available to fuel recycle reactors, kg/yr	480	—	220	—

2,020 is generated in the recycle reactors. Therefore, the average conversion ratio for this system is roughly 0.86, which again, is quite close to the equilibrium cycle value of 0.83 with U^{235} feed.

For comparative purposes, several other symbiotic systems have been considered in which plutonium was manufactured in thermal or fast reactors from U^{235} and subsequently used to provide the initial fuel to fast breeders. The plutonium-fueled fast reactors were allowed to pick up as much new capacity as they could accommodate. These systems are characterized in Table 5.2. The nuclear resource requirements for these reactor systems are shown in Fig. 5.2, together with the

TABLE 5.2. — Fuel cycle characteristics of symbiotic, 1,000-MW(e) reactor complexes

Reactor Type	Light Water	Fast Breeder	Heavy Water	Fast Breeder	Fast Converter	Fast Breeder
Fissile material	U^{235}	Pu^{239}	U^{235}	Pu^{239}	U^{235}	Pu^{239}
Fuel management	Non-Recycle	Recycle	Non-Recycle	Recycle	Non-Recycle	Recycle
Burnup, MWd/t	20,000	100,000	15,000	100,000	—	100,000
Conversion ratio	0.6	1.3	0.7	1.3	0.93	1.3
Initial specific power, kw/kg	1,000	800	3,600	800	500	800
Initial fissile requirements, (1) kg	3,900	3,700	1,040	3,700	6,100	3,700
Net U^{235} requirements, kg/yr	730	0	660	0	~800	0
Fissile Pu available, kg/yr	230	200	205	200	~800	200

(1) Includes requirements to accommodate one year external to the reactor.

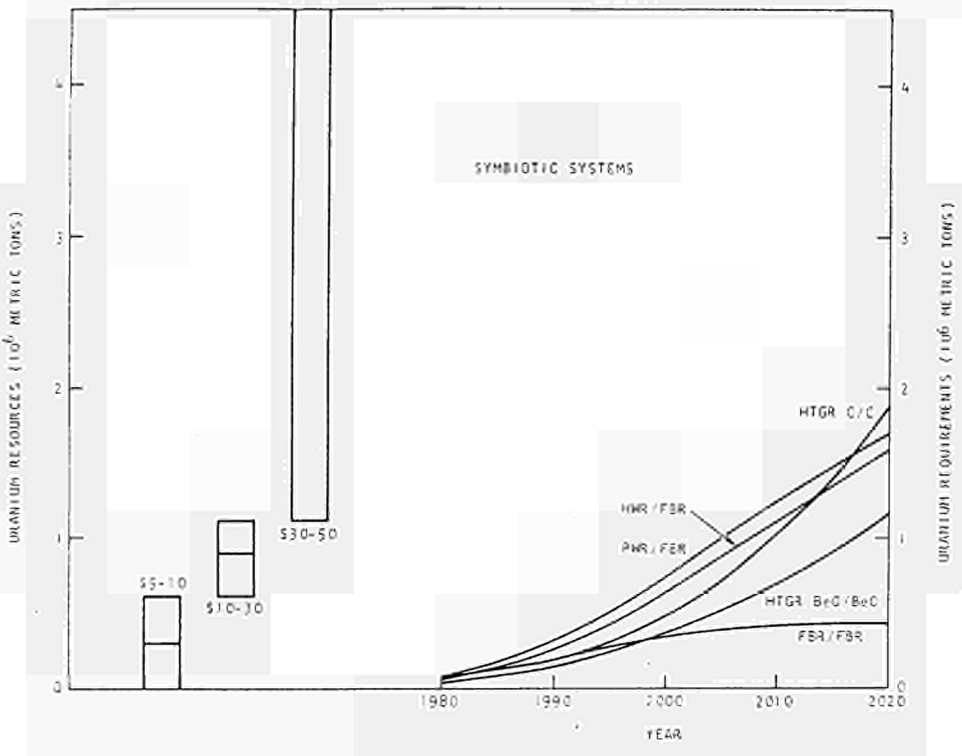

FIG. 5.2. — Uranium requirements of various reactor symbiotic systems.

HTGR systems. It is clear that, from the point of view of resource conservation only, the best way to start up the plutonium-fueled fast reactor is to use U^{235}-fueled fast reactors. The thermal reactors provide plutonium at too slow a rate. It is found that the HWR/FBR system requires about the same resources as the LWR/FBR system. The HWR uses the resources quite well, but produces very little Pu, since most of the Pu made is burned *in situ*. This behavior could be modified, but probably at a significant increase in fuel cycle costs.

The HTGR system is competitive with the U^{235} fast reactor system until the doubling time stretches out to ten years. Beyond this point the installed plutonium-fueled reactors can meet the new capacity with their own excess production of plutonium. However, it should be noted that the uranium requirements for the systems involving the FBR assume that the previously stated objectives of the FBR will be met. If, for example, the conversion ratios for these reactors should be, say, 1.1 and/or the system specific power about 400 kW/kg, because of safety and materials limitations, then all of the systems using the FBR would require in excess of 2×10^6 metric tons of uranium resources by the year 2,020. Since all of the systems involving the HTGR or the FBR show uranium requirements very close to the probable division point between low-cost and higher-cost uranium ore supplies, it is difficult to state reliable conclusions, particularly in view of the large uncertainties in avail-

able resources, nuclear energy buildup, and actual reactor performance characteristics. In any case, it is probably more significant to examine the economic performance of the various systems under various assumptions on uranium ore costs.

Tables 5.3 and 5.4 summarize the approximate fuel cycle economics calculated for the various symbiotic systems under two different assumptions on uranium ore cost. The fast breeder reactor characteristics have been chosen to be consistent with the data previously presented. For the FBR fueled with U^{235}, we have used a core that is physically similar to the Pu-fueled FBR, i.e., the fuel is oxide with about

TABLE 5.3. — Fuel cycle costs for symbiotic systems assuming fuel values based on Uranium ore at $8 per pound

Reactor System	U^{235} Cost ($/g)	(Pu^{239} + Pu^{241}) or U^{233} Value ($/g)	Fuel Cycle Cost (mills/kW-hr)		
			Feeder Reactor	Recycle Reactor	Energy-weighted Average to 2,020
HTGR/HTGR	12	14	1.1	0.95	1.0
BWR/FBR-A	12	(10)	1.6	0.90	1.2
HWR/FBR-A	12	(10)	1.4	0.90	1.1
FBR-A/FBR-A	12	(10)	1.6	0.90	1.0
FBR-C/FBR-C	12	(10)	1.5	0.70	0.7

TABLE 5.4. — Fuel cycle costs for symbiotic systems assuming fuel values based on Uranium ore at $20 per pound

Reactor System	U^{235} Cost ($/g)	(Pu^{239} + Pu^{241}) or U^{233} Value ($/g)	Fuel Cycle Cost (mills/kW-hr)		
			Feeder Reactor	Recycle Reactor	Energy-weighted Average to 2,020
HTGR/HTGR	18	21	1.4	1.2	1.3
BWR/FBR-A	18	(15)	2.3	1.05	1.5
HWR/FBR-A	18	(15)	1.9	1.05	1.4
FBR-A/FBR-A	18	(15)	2.1	1.05	1.1
FBR-C/FBR-C	18	(15)	1.9	0.80	0.8

equal fuel element dimensions and volume fractions of cladding and coolant. The specific power and conversion ratio for the fast-spectrum reactor are substantially degraded for the case where the initial fuel is U^{235}, since the spectrum-averaged fission cross section and eta values for U^{235} are considerably poorer than the values for plutonium. Consequently, the fuel cycle costs for the fast feeder reactor are significantly higher than the costs for the fast recycle reactor.

The FBR-A, it will be recalled, assumed a conversion ratio of 1.3 and a reactor specific power of 800 kW/kg for the plutonium-fueled core. The FBR-C was a higher-performance fast-spectrum reactor having a conversion ratio of 1.5. The compromise case, i.e., FBR-B, is not shown, since it was clear that it could not compete with the HTGR in the simple recycle mode.

It can be seen from the data in the tables that the thermal-spectrum reactors associated with the fast reactors all lead to poorer average fuel cycle costs than those for either the HTGR/HTGR system or the FBR/FBR systems. In addition, the fuel cycle costs individually for the LWR and HWR do not appear to be as attractive as those for the HTGR, as has been pointed out previously. Hence, in the long range, it appears that the LWR and HWR do not offer advantages as sources of plutonium for the FBR.

The fuel cycle for the FBR feeder using U^{235} fuel is quite high relative to the recycle case, but with the conversion ratio and specific power we have assumed for the Pu-fueled FBR, the integraded energy from the FBR feeder reactor over a 30-50-year period is quite small relative to that of the recycle reactor. Therefore, the high feeder fuel cycle cost in the U^{235}-fueled FBR can probably be justified in the long range on the basis of the average fuel cycle cost for the system. This argument does not apply to the thermal/fast systems since the productivity of plutonium from the thermal reactors is considerably smaller.

In conclusion, we believe that the most promising symbiotic systems are the HTGR/HTGR and the FBR/FBR systems. Looking only at the uranium conservation aspects of the various possibilities, it is apparent that the HTGR is clearly superior in performance to the other thermal-spectrum reactors by themselves, and would probably be superior to the LWR/FBR and HWR/FBR systems for the next 50 years. On the basis of resource conservation alone, the FBR/FBR system could offer advantages over all the other possibilities providing the recycle FBR is able to achieve a conversion ratio in the range of 1.3, a specific power of 800 kW/kg and a fuel burnup of 100,000 MWd/T.

Turning to the more important question of economic performance, the HTGR promises substantially lower fuel cycle costs than the LWR or HWR in the long range, and the HGTR/HGTR system can apparently offer better fuel cycle cost performance than the LWR/FBR or BWR/FBR systems for the next 50 years or more, assuming the FBR would have, on the average, performance characteristics typified by the FBR-A objectives. Although the initial operation of the FBR with U^{235} fuel would result in a relatively high fuel cycle cost, the most economic symbiotic system involving the fast breeder reactors would be the FBR/FBR.

Primarily on the basis of development status and economic potential, it would appear that the HGTR/HTGR system would gain acceptance over the other thermal reactors when the performance potential of the HTGR becomes generally accepted. If the FBR is developed to the point where the recycle reactor operation has performance characteristics typified by the FBR-A objectives, then this reactor could gain acceptance by the utility industry, providing the capital cost of the FBR plant does not exceed that of the HTGR by more than about $10/KW(e).

In summary, considering the small potential margin of improvement accomplished by the FBR-A plant over the HTGR plant, we believe that the FBR must set higher objectives. It is partly for this reason that General Atomic has focused its attention on the gas-cooled, fast breeder reactor as a long-range development concept.

We believe that it is much too early to predict the trend in future reactor acceptance with any certainty. Clearly, however, it is important that advanced nuclear power plants be available in the next few decades, particularly if the uranium ore costs should increase significantly. The HTGR has the potential for supplying the long-range energy needs of the world at economically attractive prices, even in the face of rising costs for uranium ore.

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SESSION III :

RANGE OF BURN-UPS AND CONVERSION FACTORS ECONOMIC ASSESSMENT

Introduction prepared by H. SCHMIDT, Brown Boveri/Krupp, Mannheim.

I. — INTRODUCTION

The third Session of the present Symposium is devoted to the use of thorium fuel cycles in High Temperature Reactors and the technical and economic potentialities of this system. Also the implications of the use of this type of reactor in an intermediate and long range power programme on a national or international basis is discussed.

The principal reason for the interest in this fuel cycle is the high effective eta value of the uranium-233 which is relatively insensitive to neutron spectrum changes, compared with U^{225} or Pu^{239}. Thus a good neutron economy can be guaranteed even with the hard spectrum in High Temperature Reactors which are graphite or BeO moderated. The concept under consideration can be characterised by the following properties :

(i) high conversion ratio;
(ii) high thermodynamic efficiency;
(iii) high specific power.

These characteristics are decisive for the role the system might play in a general power policy.

The following papers are submitted for this meeting :

Blomstrand, Schlösser, Bruneder, Nyffenegger, Graziani	Uranium-235/Thorium Fuel Cycles in Graphite Moderated Systems.
Carlsmith, Podeweltz, Thomas	Fuel Cycle Cost Comparisons for High Temperature Gas-Cooled Reactor Fuels.
Brandes	Uranium-235 Fueled Thermal High Temperature Reactor with Uranium-233 in Equilibrium State with Thorium-232 as the Fertile Species.
Giesser	Fuel Cycles of the "Thorium Hoch-Temperatur Reaktor" (THTR).
Dreisvogt, Schulten	Breeding in Pebble-Bed Reactors.
Wagemann	Future Fuel Requirements for Atomic Power Plants.
Stewart, Jaye, Taylor	HTGR Fuel Cycle Assessment Studies.

They can be classified into three different categories :

(i) Fuel cycles with conversion factors considerably below 1.
(ii) Breeding fuel cycles.

(iii) Future energy requirements and the changes of the High Temperature Reactor in a long-term power programme taking into account the limited amount of cheap uranium resources.

The four papers by Dragon, ORNL, Brown Boveri/Krupp (BBK) and Mr. Brandes fall into the first category. Dreisvogt's contribution comes into the second one and there are two papers, one from KFA Jülich and one from General Atomic (GA), in the last category.

The Dragon contribution discusses in great detail possible fuel cycles, once-through systems as well as fuel cycles with reprocessing. In the once-through systems a seed and blanket type arrangement with different lifetimes in the reactor is also considered. The problem of the U^{236} poisoning is discussed and a method of fuel management to reduce this problem proposed. Similar methods are applied also in the BBK-paper and have been proposed previously by GA. Fuel cycle costs are given under Dragon assumption for all the cases investigated. Of particular interest is the part where the influence of various assumptions like fabrication cost, interest rate etc. is investigated.

The paper by Carlsmith, Podeweltz and Thomas from ORNL covers a similar selection of possible cycles including low enrichment fuel at homogeneous and heterogeneous arrangements.

The work by Mr. Brandes describes a fuel cycle where fissile and fertile elements are separated with different lifetimes in the reactor. Equilibrium between Th^{232} and U^{233} is assumed. U^{235} is used as make-up fuel.

BBK describes a management scheme whereby homogeneous fuel is recycled and U^{235} is used as make up fuel. A fuel management method similar to Dragon is applied to avoid excessively high neutron losses in U^{236}.

Some of the basic assumptions made in the economic and technical assessment are given in Fig. 1. Particularly striking is the apparent uncertainty in the fuel element

BROWN BOVERI/KRUPP : COMPARISON OF ASSUMPTIONS, SESSION III FIGURE 1
Euratom Symposium Brussels, June 1965

	Dragon	ORNL	Brandes	BBK	GA
F. E. Fabrication Cost. ($/kg) . . .	420	50-200	125-500	250-1,000	100-300
F. E. Reprocessing Cost ($/kg) . .	<620	30-500	—	25-125	50-150
Interest Rate (%)	6(7)	10	—	9.6	10
Price U^{235}($/gr)	15.20	12.05	11.25	12.03	12.00
Price Th^{232}($/kg)	42	—	25	25	—
Price Graphite ($/kg)	2,8	—	—	—	6
Reactor Power (MWel)	500	1,000	1,000	250	1,000
Efficiency (%)	40	40	40	42.2	45
Power Density (MWth/m³)	10(5)	5	7	7	≈7
Specific Power (MWel/kg)	—	1,350 880	900 1,250	≈1,000	≈1,300

manufacturing cost. Most of the authors treat this quantity as a parameter to assess its influence on fuel cycle cost. The uncertainty is obviously even greater for reprocessing cost and consequently specific reprocessing costs vary still more among the various authors. The values for power, power-density, efficiency and specific power are only given for comparison and it can be seen that all the authors are in the same range of about 7 MWth/m^3 and 2.5 MWth/kg of fissile material on average.

II. — Discussion of Results

The once-through cycle, although certainly from the point of view of conservation of our energy resources not very attractive, leads to very acceptable fuel cycle cost. In addition one should bear in mind that at present this is the only cycle which seems to be technically feasible. Of particular interest here is the case with graded exposure. Dragon reports fifa values of about 1.7 with power peaks at 1.6-1.8 and or residence time of about 5 years. With their assumptions fuel cycle cost amounts to 1.5 mills/kWh for a yearly recharging system and to 1.2-1.3 mills/kWh for continuous reloading. The conversion ratio is about 0.7.

ORNL describes their minimum fuel cost case with fifa of 1.72 and a net conversion ratio of 0.72 yielding a fuel cycle cost of 1.0 mills/kWh with spent fuel discarded. In the case discharged uranium is credited, 0.885 mills/kWh is quoted. The difference in cost of 0.2 mills and the increase in conversion ratio can be easily explained by the different size of the system and different assumptions for fabrication cost and U^{235} cost.

Dragon gives also the effect of zoning on fuel cost for S about 6,000 and the once-through cycle. Complete core replacement leads to about 2.2 mills/kWh and continuous reloading to about 1.3 mills/kWh. For more than 5 zones costs vary only slowly.

A modified version of the cycle just described is to separate physically the fissile and fertile material. Dragon claims a 7-8 % increase in conversion ratio and a 10 % increase in fifa. Difficulties concerning power peaks have to be overcome. Dragon proposes a residence time of the blanket three times the one of the seed. It is obvious that this fuel management scheme has an advantage if the fabrication costs are high, since it allows a higher residence time of a major part of the fissile and fertile material. The maximum improvement one might expect is in the order of 0.2-0.3 mills/kWh depending on fabrication cost.

In the Dragon study, a case with a fertile blanket with residence times comparable with the reactor life, is investigated. The seed elements are to be recharged frequently. Average fuel costs are expected to be around 1.3 mills/kWh.

The fuel cycle with reprocessing of the discharged fuel and U^{235} make-up has been treated by the first four papers. The absorption in higher uranium isotopes which will not be removed during reprocessing presents the main difficulty. Dragon and BBK use, as mentioned above, a particular fuel management and avoid in this way excessive U^{236} build-up. ORNL obtains because of this phenomenon a somewhat

unfavourable fuel cycle. In the work by Brandes the U^{233} chain ends already with U^{234} although equilibrium is considered. The results of Dragon and BBK for about \$500/kg fuel element fabrication and reprocessing cost, an S-value of 6,000, and fuel lifetimes for 5.5 and 4.5 years lead to about 1.2-1.3 mills/kWh in the Dragon assessment and to 1.0 mills in the BBK-paper. The difference is apparently due to the fact that BBK costs do not include interest on fuel in the reactor.

The ORNL results cannot be directly compared, because of different assumptions. Here the fuel cycle with reprocessing leads only to a small improvement giving 0.880 mills/kWh instead of 0.885 in the once-through case. It should be born in mind that ORNL results refer to a very large system.

Considerably lower fuel cycle costs are given in the paper by Brandes with 0.65 mills/kWh even with fabrication and reprocessing charges of \$500/kg and with a specific power of about 2.4 MWth/kg. This is partly due to the neglection of interest rates for fissile material and fabrication cost and also due to the favourable fuel management.

Breeding in a High Temperature Reactor is discussed by Dreisvogt and Schulten. In a somewhat unusual type of analysis they investigate to what extent breeding is possible in these reactors. They are using, like Brandes, a two type element concept for fertile and fissile material. Already from the start a selfsustaining system with Th^{-223} and U^{-233} being in equilibrium is assumed. The fissile and fertile element lifetime is estimated and optimized afterwards by evaluating the fission product poisoning. It is surprising that even at relatively high burn-ups, more than 30,000 MWd/t, breeding is possible. The given doubling times have to be looked at in the light of the very fast reprocessing plant adopted. Only 10 % of the fuel is outside the reactor.

In an unpublished paper Schlösser has performed some burn-up calculations using as fissile material segregated fuel and thorium as fertile material. He shows that in a large Pebble Bed reactor — a neutron loss of about 1 % was assumed — an overall conversion factor of one can be achieved for a burn-up of about one fifa, or for fuel residence times of about 3 years. All fission products are retained in the core and no beryllium is used. Obviously for a smaller burn-up a certain amount of breeding can be obtained. For instance, for a burn-up of only 0.5 fifa the overall conversion factor is about 1.04. The specific power is around 1 MWth/kg, and the average power density 5 MW/m^3. In these calculations a complete utilisation of the produced protactinium is assumed.

The conversion factors quoted refer to a cycle, whereby the uranium isotopes U^{233} to U^{236} are in equilibrium. According to the residence time and the breeding gain the equilibrium isotopic composition varies. For a high breeding factor about as much U^{236} will be present as U^{235}, whereas for a breeding factor near one the fuel will contain about twice as much U^{236} as U^{235}. For this reason it is necessary to start off this cycle with segregated fuel as otherwise the fraction of U^{236} atoms to fissile atoms is too high and breeding at reasonable burn-ups is prevented.

The question about the possible role of the HTGR in a long term power programme is discussed in the last two papers.

Wagemann, KFA-Jülich, describes several combinations of systems, generally a converter followed by a breeder. Four different models are adopted, constant conversion ratio with a step change from converter to breeder, a slow change-over from converter to breeder, and a continuously increasing conversion ratio. Specific power, conversion ratio and doubling time are used as parameters.

The main result is, that not only conversion ratio and doubling time are important, but also specific power. This is generally the case in a rapidly expanding power production. The power demand estimation of the USA was taken as reference.

The paper by Stewart and *al.* from General Atomic investigates the role of the High Temperature Reactor in this context as well. They arrive at the same conclusions and find that the system is very well suited to satisfy future power demands considering the limited amounts of cheap uranium and the power forecast of the USA. They stress further two points which are essential and which are met by the HTGR-system. These are :

(i) high thermodynamic efficiency;

(ii) reasonably long residence time of the fuel in the reactor compared with the reprocessing time.

In comparison with the Light and Heavy Water Reactors the HTGR has the lowest fuel requirements up to the year 2,020.

The influence of ore cost on fuel cycle cost is studied for the three converters LWR, HWR and HTGR. Here again the HTGR compares favourably with the two others. Taking 1.7 mills/kWh as a critical fuel cost — to be competitive with fossile fuel power stations — critical ore cost for LWR would be at about 10$/lb, for HWR at about 15$/lb, and for HTGR higher than 40$/lb. It is this characteristic which puts the HTGR in the position to use the large uranium and thorium resources available at recovery cost between 30 and 50$/lb. The estimated cost for recovery of uranium from sea water is well in the range for economical use in an HTGR. Changing ore cost from 10$/lb to 40$/lb would lead to an increase from 1 mills/kWh to about 1.6 mills/kWh.

III. — Conclusion

A considerable amount of material has been presented about this system, which is still under development.

Clearly, events of the next years will provide a much improved basis for the evaluation of the High Temperature Reactor Fuel Cycle. Three reactors will be operating and one can expect also a much better knowledge of fabrication and reprocessing problems. Generally the impression prevails, although direct comparison is difficult, that there is good agreement in the neutron physics calculation even with

the different methods used. It is because of the uncertainty and the differences in the technical and economic assumptions that the results vary considerably.

However, on the basis of an interest rate of about 8-10 % and for a reactor of the size of about 500 MWel with 40 % efficiency and 7 MW/m^3 power density one would expect the following range of fuel cycle cost, with fabrication and reprocessing charges of about $400/kg in the cycle with reprocessing and $300/kg in the once-through cycle.

The once-through cycle with no credit for discharged uranium for a continuous loading scheme would lead to about 1.3-1.4 mills/kWh; a credit of about 0.1-0.15 mills/kWh might be expected.

Fuel cycles with reprocessing lead very likely to only a very small improvement in cost although from the point of view of efficient utilisation of energy resources they are much more favourable. Cost of 1.1-1.2 mills/kWh would perhaps be typical for this cycle.

A separation of fissile and fertile elements leads to an improvement in fuel cycle cost. If there are no restrictions on power peaking and burn-up, up to 0.3 mills/kWh can be gained.

The long term power need can be met by the system under consideration, because of good neutron economy and high specific power. In this context it is particularly important, that the fuel cycle costs are relatively insensitive to ore costs.

Generally, the prospects of the cycle are good and the final success will depend upon development of an inexpensive fuel fabrication and recycle technology.

SESSION III : DISCUSSION

Chairman : P. CAPRIOGLIO (Euratom)

1. — NEUTRON CROSS SECTIONS AND FISSION YIELDS

Dr. CAPRIOGLIO : There are a few questions that have been raised in Dr. Schmidt's presentation which come to my mind immediately and to start with an easy one, I am wondering what the situation is with the neutron data which we have now. Is there a sufficient agreement on them ? Are the cross sections of protactinium in various spectra known well enough to be satisfactory ? Are the fission products cross sections known in a sufficient way to make calculations for very long burn-ups satisfactory enough ? And how does this compare in the two cases of once-through and reprocessing and how these uncertainties affect conversion factors, if one tries to go to breeding ?

It is my feeling that if one accepts as a goal to make cheaper electricity, perhaps one would be rather satisfied with the data one has today. If, on the contrary, one wants absolutely to go to breeding, and we will have plenty of discussion on this, I am sure, later on, one would perhaps like to have more precise information available. I am wondering if anybody wants to comment on this ? Would you like to start, Dr. Stewart ?

Dr. STEWART *(General Atomic)* : While you really asked several questions, let me try to answer these simultaneously, and maybe the best way to go about this is to look first at some neutron balances which I have. So, could I have the first slide ? This slide is a table (no 4.4) that appears in our report with the conference. It shows the neutron balances that one might be able to obtain under various kinds of operating conditions. In the first case, we have assumed that the reactor is fuelled with U^{235} initially, that the moderator is completely graphite and that the fuel lifetime is 4 years.

In the first four cases that we have looked at, we have assumed that we have used coated fuel particles so that the volatile fission products are completely retained. Now, if one looks at the bottom part of the table, one can see the neutron losses to leakage, to the graphite, to protactinium, to the U^{236} and neptunium-237, to fission product poisons (we have separated xenon-135 out from the other fission product poisons) and finally the losses to control.

The second column is a rather academic case where one assumes that after one cycle of operation, one takes the U^{233} that has been generated and starts up subsequent reactors with U^{233} only. In practice, of course, the fuel would consist of both U^{233} and U^{235}. The thing that is interesting is that even with this very optimistic case where the fuel is completely U^{233}, the conversion ratio that we calculate is at best something like 0.99. This assumes a 3 year fuel lifetime.

For the more realistic case, where one assumes an equilibrium build-up of the U^{233}, (incidentally, we have used a fuel management programme, which recycles the bred fuel once through the reactor), you see that with a reactor using an all graphite core, the calculated conversion ratio is about 0.87.

In the fourth case, where we have assumed that the spines of the fuel element are replaced by beryllium oxide, we obtain a somewhat higher eta-value because of the softer spectrum, and we get some multiplication due to the $(n, 2n)$ effect in the beryllium. With a 4 year cycle here, we would calculate a conversion ratio of 0.99. Only in the case where one is willing to withdraw the volatile fission products, particularly the xenon-135, could one get conversion ratios substantially above unity.

Now let me point out a couple of things about the table. First of all, we have not assumed any thorium blankets for the reactors, although I believe in case 2, we did assume that there was a thorium blanket. One could improve the conversion ratio perhaps by 0.02 by surrounding the core with a thorium blanket. This is indicated by the difference in leakage between the case 2 and the others. One could improve the conversion ratio by some sort of an on-stream refuelling, thereby eliminating control. This would pick up something like 0.03 as you can see from the tables. Beyond this, it does not look like there is much room to improve conversion ratios, although we feel that the conversion ratios shown here are quite good.

Now, let's come back to one of the other questions, namely how does the uncertainty in cross section data affect the performance characteristics of the HTGR. You can see that the total loss in conversion ratio from the protactinium absorption is in the neighbourhood of 0.03, so that an uncertainty in the cross section of protactinium of say 50 % would only amount to 0.01 in conversion ratio. The fission product poisons, other than xenon, altogether represent something like 10 % in the conversion ratio. An uncertainty in the fission product poison cross sections of say 10 % or even 20 % would only amount to something like 0.01 or 0.02 in the conversion ratio. Note, however, that if one loses 0.01 in conversion ratio, then one has to use more U^{235} in the make-up to account for the difference between unity conversion ratio and something less than unity. When one has to use more U^{235}, the spectrum average value of the eta for the U^{233} and U^{235} is somewhat smaller; and our calculations show that if one loses say 0.01 directly from some neutron balance loss, the net-effect in the recycle, which comes about because of the larger requirements for U^{235} make-up contributes another loss of about 0.01. So, when I say that an uncertainty in any of these cross sections might amount to 0.01 or 0.02 in conversion ratio, one has to remember that this has to be doubled approximately when one takes into consideration the recycle.

The biggest source of uncertainty, we would think, might come from the uncertainty in the eta-values themselves. Until about one year ago, there was a large uncertainty in the U^{233} eta-value. Some sources quoted an epithermal value of eta for U^{233} integrated from 1 eV up of about 2.14, while other sources were as low

as 2.00. This, as you can see, would make a big difference in the conversion ratio, even when you take into consideration that most of the absorptions are in the thermal range. This turns out to lead to an uncertainty in conversion ratio of perhaps 0.05 or maybe even larger. More recent integral experiments have indicated that the 2.14 epithermal resonance integral for eta is the better value, and therefore all our calculations have used the more optimistic eta-value. But one must keep in mind that if subsequent experiments should show the eta-value for U^{233} not to be so optimistic, this could compromise the conversion ratios slightly.

I think that is about all I want to say then about the uncertainty on cross sections and I apologise for taking so much time, but I would like to say something about some of the implications of these things now and I have a couple more slides I would like to show. If I could go to the next one, this shows the difference in conversion ratio that one would get between the all graphite core and a core using a beryllium oxide spine in the fuel elements (fig. 4.5). The conversion ratio is plotted as a function of the fuel lifetime, that is the number of years that the fuel is left in the reactor. As I recall it, a fuel lifetime of around 4 years correspond to a fifa, i.e. fissions per initial fissile atom, of a little bit over 1.0. Now, one can improve conversion ratios significantly by going to short fuel lifetime, as Dr. Schmidt has indicated in his review. But one does this, as we all know, at the expense of fuel cycle cost.

My next slide (fig. 4.4) illustrates how the fuel lifetime affects the fuel cycle cost if one makes various assumptions on fabrication and recovery costs. As one would expect, for a very low fabrication and recovery cost, one can afford to operate the fuel cycle for a fairly short fuel lifetime, and one can see, that even going back to 3 years fuel lifetime, the penalty is not very great. The optimum fuel cycle is about 4 years for this case. On the other hand, if one has to pay 300 dollars per kilogram for the combination of fabrication and recovery, then the optimum fuel lifetime is more like 5 1/2 years, and you see that there is about a 0.2 mills/KWh penalty associated with the higher fabrication and recovery costs. If one has to go to a combined cost of 450 dollars per kilogram for the fabrication and recovery, then the optimum fuel lifetime is more like 7 years and we lose another approximately 0.2 mills/kWh in the fuel cycle cost. You can see, therefore, that it is very important for us ultimately to be able to get to quite cheap fabrication and recovery costs.

Dr. CAPRIOGLIO : I think this is an extremely important subject, as Dr. Stewart has pointed out, and I would like to have a short discussion on this before asking the other authors to comment on Dr. Schmidt's presentation.

Dr. SANDERS *(UKAEA Winfrith)* : Could I ask Dr Stewart for his views on the accuracy of the enhancement one can expect from the beryllia spines in the elements and in particular, how has the neutron loss by the (n, alpha) and the lithium 6 been accounted for in these calculations? Certainly, until recently, data on the (n, 2n) reaction in beryllium have not been very well founded, and I wonder whether he has got experimental evidence to justify these increases in conversion ratio that one obtains with the beryllia spine ?

Dr. STEWART *(General Atomic)* : You recall that there was something like a 0.10 difference between the all graphite core and the core containing some beryllium oxide. Incidentally, the volume fraction of the beryllium oxide relative to the graphite is something like 30 % in these cases we have looked at. About half of the effect or half of the difference between the graphite and the beryllium oxide cases comes about because of the (n, 2n) effect, and the other half due to a better spectral softening effect from the beryllium oxide. So that actually the (n, 2n) effect itself, minus the losses to the (n, alpha) and the lithium, only amounts to perhaps 0.05 in the conversion ratio. The effective epsilon value that we indicate, corresponds to the (n, 2n) minus the (n, alpha) effects. We have put the neutron absorption in the lithium in the moderator absorption. If one refers to the table, the moderator effect can be seen to be about 0.01, or perhaps a little bit more, which comes about because of the absorption in lithium. We, of course, assume that after each recycle the beryllium oxide is reprocessed and the lithium is removed. As to the accuracy of the (n, 2n) and the (n, alpha) cross section data, I cannot quote you exact sources. Mr. Jaye, who is here with me, may be able to shed some light on this, but we have used the most recent differential data that are available for these cross sections, and we believe that the uncertainty in this probably would not amount to more than say 0.02 in conversion ratio. Would you agree ?

Mr. JAYE *(General Atomic)* : Yes, I am afraid, I also cannot recall the sources entirely, although I think they are primarily out of the BNL 325.

Mr. BLOMSTRAND *(Dragon Project)* : In this connection I should like to mention that for the fission products it is not only important to know the cross sections but as well the fission yields. Several fission products have such high cross section that they absorb neutrons quite quickly after having been formed, hence these losses depend more on the fission yields than the cross sections.

These yields are spectrum sensitive. In the fuel cycle calculations we normally assume the values for the thermal yields but if there is a considerable amount of epithermal fission the overall yields may be different. This might influence the fuel cycle analysis somewhat.

I should like to make another comment to what Dr. Stewart said on the neutron balance in these systems. He mentioned that if the neutron losses can be decreased in a system using recycled fuel this increases the U^{233}/U^{235} ratio in the reactor which improves the overall η-value and raises the conversion. Now, we have seen that for the most important fission products the yields from U^{233} are lower than for U^{235}. This means that the higher the U^{233} content in a system is, the lower are the fission product losses which helps in addition to put up the conversion.

Dr. STEWART *(General Atomic)* : The difference in yields for U^{233} and U^{235} have been taken into consideration in the calculations that were indicated. Insofar as the uncertainty in yields, of course, one must remember that approximately 85 %, as I recall, of the fissions occur below something like 1 eV, so that even if

there is a fairly large uncertainty in the yield in the epithermal energy range, this is not going to have a terribly significant effect, I believe, on the overall fission product poisoning.

Mr. JAYE *(General Atomic)* : There is just one other comment on the yield. In the fission products that are most significant, I believe it is the promethium-147, 148 and on up chain, this turns out to be on the dropping side of the high mass peak, and while the uncertainties are fairly great there, that is not the region that is affected by a change in fission spectrum. It is generally the dip between the two mass peaks.

Dr. DE NORDWALL *(AERE Harwell)* : Could you make a comment, or could somebody make a comment on the effect of silicon carbide on the neutron economy in the fuel cycle cost?

Dr. CAPRIOGLIO : Thank you for this question. I would like however, to take it up further on, if you do not mind. If possible, I would like to keep the discussion in order and would like to restrict now to this problem of cross section and availability of enough information on nuclear data. Any further question on this? I have one myself which is somewhat naive. What is the economic consequence of this uncertainty that all of you are talking about? I appreciate that there is a large difference between having a nice doubling time or practically no breeding at all. We will come back to this later on. But on economic grounds, assuming that we are only interested in making cheaper electricity for the moment, is there any significant importance of these uncertainties and is there any serious incentive to have more precise data or not?

Dr. STEWART *(General Atomic)* : The effect of changes in conversion ratio, the size we are talking about, is quite small in the fuel cycle cost. As I recall it, and I was just checking this with Mr. Jaye, a change of 0.10 in conversion ratio would make a change in fuel cycle cost of something like 0.15 mills/kWh and we are talking of 10 % of that.

Dr. SCHROEDER *(Dragon Project)* : This comment by Dr. Stewart is valid for the present economic situation but, if there were a rise in fissile material costs, that would of course reflect more through higher conversion ratios.

Dr. CAPRIOGLIO : Yes, I think this is quite clear.

Dr. HINTERMANN *(Dragon Project)* : When we speak about the economy of recycle or once-through, we find that fuel once-through gives already reasonable fuel costs. Therefore, we can afford refabrication and reprocessing costs which are up to the new fuel costs. If we speak of the once-through cycles, then the conversion would not be of interest at all, but only the fifa. Therefore, we cannot say, we must be reasonable in conversion. It was mentioned by Dr. Schmidt and Dr. Stewart that we should go towards cheap reprocessing cost and cheap refabrication

cost. Now, if we start reprocessing, it is realistic to assume for the beginning possibly quite high processing costs; this means we can afford as much as new fuel costs. Cheap reprocessing and refabrication is not a must but it gives a good credit and makes conversion interesting.

Dr. GRATTON *(UKAEA Winfrith)* : Although the burn-up effects and breeding etc. are very important to the long term economy of our reactor, we must recognize that U^{235} will play a very important role in getting to the thorium-233 cycle with the high conversion factor. I would like to be brought up to date on what the state of our design calculations are in predicting the reactivity of a clean U^{235}-thorium reactor ?

Prof. SCHULTEN *(KFA/THTR)* : I believe, so far as high conversion factors and breeding factors are considered, the most important nuclear data is the eta value. Because when we will go to unity, then in any case the contribution of fission products must be very small so that the error which we are making in the fission products poisoning, when we are in the order of plus or minus 10 %, this error for the whole balance is not so important. Also the influence of the protactinium cross sections is very important for the considerations for these cases, where high specific power is considered. But as I know, the cross section for protactinium is now measured very well, and I think there is no doubt that the data for protactinium are very good, so that the real lack to say something more exact about the breeding factors or about high conversion factors, is really the eta-value.

Dr. SCHROEDER *(Dragon Project)* : Concerning the accuracy of predictions of reactivities, or in other words, how much margin must one leave for the uncertainty in reactivity predictions? We have just loaded Dragon and compared our predictions with experimental results; and, although we have a difference of 3 % reactivity, this does not mean this is the order of magnitude you should expect in a large reactor. I would like to recall that Dragon is a very small unit with a leakage of over 50 % with a very odd control rod configuration, and to hit reactivity within 3 % is quite good, where we have an excess reactivity of more than 24 %. Apart from that, the experimental figures we are comparing and theoretical figures calculated are not quite the same. As far as the feedback on the economics of large systems is concerned, I think this is very small and we can be confident in the methods so that we must not leave a margin which is larger than a percent or so.

Mr. BLOMSTRAND *(Dragon Project)* : I would like to mention that the only cross section in these fuel cycle calculations that we do not need to know very accurately for making cost optimisations, is the thorium cross section. If we are a little wrong in the thorium cross section, this will only lead to too high thorium content in the system: but it will still give correct conversion ratios, because the conversion ratio is only defined by the amount of excess neutrons that are not absorbed in other isotopes. If one wants to make as Mr. Gratton suggests, a reactivity prediction on the core which is loaded with uranium and thorium, then one is left with the

problem of calculating very accurately the resonance absorption in thorium. This is something that does not really come in, when one makes survey calculations on the thorium cycle.

Dr. STEWART *(General Atomic)* : Just to reinforce some of the statements already made on the question, I guess they were made by Dr. Sanders, we have looked at a number of critical experiments using U^{235} and graphite, including one that we did ourselves at the laboratory, and we feel that we can calculate these assemblies to something like 1 % to 1 % in K's, so that on the basis of this we believe that we know our U^{235} cross section data quite well.

Mr. FORBES-GOWER *(CEGB)* : I was very interested by Mr. Schröder's remark that he was out by 3 % in reactivity on a reactor calculation. Now admitting that he has a very difficult case and a small core with a very high leakage, we are in fact considering small cores because the whole attempt to reduce capital charges goes in the direction of small cores. We shall forever have to consider fairly high leakage systems and what Mr. Schröder's remark indicates to me, is that the reactor calculation is as important as the reactor data. And the effect of being out in a reactor calculation by a few percent, while having been lightly dismissed, is very great, because it would alter the lifetime of your charge, it would alter the interest rates, it would alter the optimum moderator ratio for any given case, this in turn would alter conversion ratios and everything else. I feel that one ought to consider, at the same time as data accuracies, the effect of the reactor calculations since so many of the fuel cycle costs being presented here, have not had very good reactor calculations going with them.

Dr. SCHLOESSER *(Euratom/THTR)* : I think one should not mix small cores with high leakage. I mean even the large system as CEGB has at the moment, have a leakage which is relatively large. In the small reactors we are discussing here, the loading is heavy so that the leakage is relatively small. In the case of Dragon we should not compare the small size but the high leakage which is 50 %. It would be about 3 % in a large power reactor. In this way the uncertainty in the calculations is reduced. Furthermore, it should be considered that the control rods are in the reflector. There the flux, the thermal flux especially, changes rapidly. In a power reactor control rods would be in the core, with hardly any change in the thermal flux and the uncertainty in the control rods calculations would be reduced as well.

Dr. SCHROEDER *(Dragon Project)* : I have just a very short remark. There is some economics in Dragon as well and the 3 % I am talking about, amounts to a difference in expected lifetime of plus or minus 10 days, compared with the total lifetime we expect of 200 days. So, you see there is only 5 % uncertainty in the total lifetime resulting from the uncertain calculations.

Dr. SANDERS *(UKAEA Winfrith)* : Returning to the question of the accuracy of reactivity prediction, the experiments that we made in the Zenith reactor (which is similar in dimensions to Dragon) over a range of carbon to U^{235} ratios from 1,000

up to 8,000 showed that one could calculate the reactivity of this system to accuracy of about plus or minus 2 %. These were one zone core which I think present a rather simpler problem than the Dragon reactor itself. I think our conclusions tend to bear out Dr. Stewart's comments on the results of the G. A. critical experiments that this is the order of accuracy which one can expect on the present nuclear data.

Mr. JAYE *(General Atomic)* : I should like to make just one remark about the comment that the calculations referred to in the papers being presented were quote "not too good". I do not really think this is true. On the one hand, most of the papers considered point reactor type calculations which for very small cores like Peach Bottom or Dragon would be quite questionable. But the authors were really considering very large size reactors and very small leakage systems and I know our calculations at least have been checked out with dimensional calculations and have been shown to be quite accurate. So I really believe that the calculations as presented in the papers this morning are not as bad as they would look if one were to try to apply them to the wrong size systems.

Dr. CAPRIOGLIO : It seems to me that a certain confidence is shown by the designers about their calculations and this is very comforting indeed.

Mr. FORBES-GOWER *(CEGB)* : Could I just take up one small point on this reactor calculation. Mr. Schröder said that he only gets an alteration of a few days in Dragon, but Dragon surely is not a system with a very high conversion factor. With a high conversion factor must go a very flat reactivity curve or a fairly flat reactivity curve. The moment you get a fairly flat reactivity curve 1 % in reactivity represents quite a large change in reactor lifetime, if you are on a reactivity limit.

Dr. CAPRIOGLIO : This sounds very convincing to me. I would like to have one answer only and since it is Dragon who is asked, I would like to see if anybody from Dragon wants to give a last answer.

Dr. SCHROEDER *(Dragon Project)*: It is right that the reactivity curve is quite flat in the big systems because we have the high conversion ratio. If you leave a margin of a percent and you are right whatever the uncertainties you expect, then that margin is preserved for a long time due to the flat reactivity curve and the same argument applies.

2. — AUTHOR'S COMMENTS

Dr. CAPRIOGLIO : I would like now to call for various authors to make comments on Dr. Schmidt's presentation.

Mr. BLOMSTRAND *(Dragon Project)* : In addition to the comments. Dr. Schmidt has made on our paper, I should just like to stress a few points. The most important assumption that we make when we do these calculations is that of 3 % leakage. We have got this figure from an engineering design study that was made on a Power Station with a 1,250 MW thermal output. If one can cut these leakage losses, then one can gain considerably in fifa and conversion ratio. Other assumptions that one makes such as voidage, fuel temperature, moderator temperature, would in-

fluence a little the nuclear design in terms of N-values and S-values but they would not change the optimum fifas or the optimum conversion ratios.

The next point that I want to make concerns the limits on specific power density and absolute power density that are introduced by protactinium and xenon poisoning. I think that the main absorption in proctatinium lies m the resonance region and this is much more important than the thermal poisoning. Thus, if one tries to put up the absolute power density in the reactor, then one puts up the epithermal flux which increases the protactinium poisoning. We think that the protactinium poisoning puts a limit on absolute power density rather than on specific power density. The specific power density starts to be limited by thermal protactinium poisoning only when one goes to very well moderated systems but here the xenon poisoning is more important, it increases strongly if one increases the specific power density. Otherwise the physics parameters feed back very little on the nuclear design provided that one has an S-value (that is a carbon to uranium ratio) in the average system higher than 4,000. They feed back very little on fifa and conversion ratio, but they do feed back on the N-value. The reason for this is that, whether one has a well moderated or an under-moderated system, the number of neutrons available for conversion are roughly the same. But in a well-moderated system, one cannot utilize the thorium resonances for conversion and hence one has got to put in more thorium to catch all these extra neutrons in the thermal region. Consequently the difference between under-moderated and well-moderated systems is the higher N-values of the well-moderated systems. Now this comes into the cost calculations. If we assume very high charges for reprocessing and fabrication, then we have to take the penalty in well-moderated system of the higher turn-over of thorium through the fabrication and reprocessing plants. So, if there was no such thing as interest, we would like to go to an under-moderated system with a low N-value. Unfortunately, in an under-moderated system, one has very high inventory of fuel and one has to pay quite a high amount of interest on that, so the optimum between an under-moderated system and a well-moderated system are balanced on one hand of the high interest costs on fissile inventory in the under-moderated systems and the higher turn-over costs of heavy metal through reprocessing and fabrication plants for well-moderated systems. It is quite interesting to compare how different assumptions really feed back on nuclear design. I have mentioned that if there was no interest, we would like to have an average S-value of perhaps 4,000 in the core. We have considered an interest rate of 7 % and charges for fabrication and reprocessing to be in the order of 100 pounds sterling per kilogram heavy metal independent of throughputs. This gives us an optimum S-value of in the order of 6,000 or 7,000. We would not like to go lower than that because then we would lose too much in interest on the inventory. I would just like to mention that Mr. Carlsmith has made virtually the identical nuclear analysis as we have done. He has assumed 10 % interest rate on the fuel inventory and has assumed throughput fabrication and reprocessing charges so that if he goes to higher throughput, his charges per kilogram are going down. On the same cycle — I refer now to the once-through

cycle, no reprocessing and uranium/thorium — he ends up with an average S-value of 13,000, which implies twice the specific power density that we would consider. These differences all depend on his different cost assumptions. I think that if one reprocesses, then the part of the total fuel costs, which is dependent on the working capital charges on the inventory, is lower, and the part consisting of heavy metal turn-over is higher; consequently there will be an incentive to go to slightly more under moderated systems on reprocessing cycles on once-through cycles.

The next thing I want to talk about is the feed back of power density on the conversion ratio and in that connection I would be interested to hear later from Dr. Stewart what power density and specific power density he was assuming in the slides and curves that he was showing. We find on the once-through cycle that the protactinium poisoning has a very important influence on the conversion ratio because, as Dr. Stewart showed, it goes in with the factor two in the neutron balance. Every neutron lost in protactinium means, that the neutron that was absorbed in thorium for production of that particular protactinium atom was really non-fertile. If we have a power density of 5 MW per m^3 on the once-through cycle, we get to conversion ratios of 0.8 At 10 MW per m^3 we are down to 0.7; if we go up to 15 MW per m^3 we are down in the conversion ratio to almost 0.6, so I think this is quite a strong dependence.

If one then looks at the fuel cycle costs for different power densities in the once-through cycles, one comes to the very interesting conclusion that what one loses in U^{233} production when one goes to higher power densities due to the burning of protactinium, one gains in interest charges on the fuel inventory due to the higher specific power. Indeed we find that provided that the S-value of the system is larger than 6,000, it does not matter for the fuel cycle cost what power density one uses between 5 and 15 MW per m^3. Now, this is only true for the once-through cycles. I would have liked to report how the power density influences the costs for the reprocessing cycles but they are a little more complicated and more fuel management schemes have to be studied. We believe that there is an optimum towards the lower power densities in the order of 5 or 7 MW per m^3.

Concerning the power peak in the fresh fuel, I can mention that it depends on the fabrication charges, because if we can go to lower fabrication charges on the once-through cycle, we are also allowed to reduce the fuel element residence times. We find that a reduction in the fabrication charges within 50 pounds sterling per kilogram corresponds to an improvement of 10 % in the power peak. On reprocessing cycles, assuming a power density of 10 MW per m^3, we find that the U^{233} recycle increases the conversion ratio from 0.7 on the once-through cycle to around 0.85 on the segregated fuel cycle. These comes from two effects : Dr. Stewart has mentioned the higher eta values in these systems because of their higher U^{233} content mentioned earlier that it is interesting that the lower U^{233} yields for the fission products help in the same way to push up the conversion ratio. We have found that we get the same fuel costs for the once-through cycle and the reprocessing cycle on segregated fuel, if we assume a reprocessing charge of 200 pounds sterling

per kilogram heavy metal. I want to stress that this figure includes the remote refabrication of fuel. (One has to have remote refabrication because of the activity of U^{232}.)

I have an extra comment to make : If one wants to compare fuel costs, we quote them in pence per kilowatt/hours and the American papers quote them in mills per kilowatt/hours. The conversion factor between mills and pence is 70 over 6. Now, we have assumed 14 dollars per kilogram for U^{235}; in the American papers the same figure is 12 dollars per kilogram. Thus if one takes our figures in pence per kWh and multiplies them by 10, we will get the fuel costs in mills/kWh calculated under the assumption of 12 dollars per kilogram U^{235} charge. This is just chance but gives an easy way of comparing the results.

Dr. CAPRIOGLIO : I think that Mr. Blomstrand's remarks have now given us plenty of scope for interesting discussion. There were a few challenging questions put forward such as that the *protactinium loss* is not depending on specific power which sounds new to me and I would like to find out, if there is any question on this.

Mr. JAYE *(General Atomic)* : I think, we generally agree with Mr. Blomstrand that the protactinium losses are not universally associated with a specific power but are rather dependent on both the specific power and the power density. The evidence has been from the measurements at the MTR by Simpson that the resonances seem to occur below 10 volts; at least the majority of the resonance integral is there, and therefore it is in a very difficult region to assess for an HTGR. This is just the area between 1 and 10 volts where you wonder whether you are talking about a thermal spectrum or an epithermal spectrum. It is not either the slowing down or thermal energy range; it is an intermediate range, and I think, therefore, you must find that you cannot be exclusively looking at specific power or exclusively looking at power density; you must look at the whole picture at one time.

Dr. CAPRIOGLIO : As a matter of terminology, and in order to be sure that we understand each other, we talk about power density as volume power density and about specific power as the rating of fuel.

I had a question myself concerning this matter of *specific power and power density*. I assume all these optimisations have been made taking into account only the fuel cycle cost. There must be some feedback from the capital cost, if one goes to very low power densities, so even if for pure fuel cycle calculations, one is tempted to go to very low power densities, there are certainly other considerations which could be important that would tend to push up the power density again. May I ask Mr. Blomstrand of this has been taken into account and what he feels about it?

Mr. BLOMSTRAND *(Dragon Project)* : I think this is a very valid point and one that we fuel cycle people do not really look into. We would like very much to do it, but it is a matter of time and effort. Certainly, if one goes to lower power

density, this would imply a larger station and larger core sizes because one would like to keep the total electric output of the system constant. But how much this feeds back on capital cost, I just cannot say. I think Mr. Lockett would like to say a few words.

Mr. LOCKETT *(Dragon Project)* : Yes, it is quite true that there is a significant influence here, but while we are talking about say a difference between 10 MW and 5 MW per m^3 the effect on a large size reactor is not as important as you might think, because the core size itself relative to the reflector thickness plus heat exchanger, space and so on, does not make the pressure vessel so much larger and can, by making things as compact as possible, usually succeed in going as far as possible towards the preferred nuclear optimum without spoiling the capital cost side of the story. At present, we seem to have jumped midway between 5 and 10 MW per m^3 as something like a preferred optimum between physics and engineering size. This is perhaps all I can say, but I agree that it does amount to rather careful design to try and mix the two to the best advantage.

Mr. CARLSMITH *(ORNL)* : I think there is another point that also has a bearing on this question of trying to increase the power density and that is that one normally finds that if an effort is made to increase the power density much above the levels which have been indicated in these designs, that there is difficulty in the thermal design of the fuel elements. The temperatures in the fuel elements either become excessive or else one is forced to very small fuel elements which are increases of cost so that for this reason also I think there has not been too much incentive to try to investigate designs which would reduce the capital cost slightly by increasing the power densities.

Mr. JAYE *(General Atomic)* : There is just one comment I would like to make on some of these very large S-values that have been considered by Dragon and Oak Ridge. I wonder, if they would like to comment on how these might change, if they include the cost of the core aside from the cost of the fuel particles. That is, the graphite which is almost fixed in cost and for a given fifa and very large S-value results in a small amount of energy to be amortized over.

Mr. BLOMSTRAND *(Dragon Project)* : I am not sure, if I understood the questions correctly. We include the graphite costs in our calculations. Certainly, if one goes to higher S-values, this means a quicker turn-over of graphite because one gets to shorter fuel elements residence time but we have seen that up to an S-value of at least 10,000 on average this is not really serious. We have assumed 1 pound sterling per kilogram graphite and I hope it can be manufactured for that.

Dr. SCHMIDT *(BBK/THTR)* : There was just one more comment as far as the power density is concerned. Mr. Carlsmith was just mentioning the effect of thermal limitations of fuel elements. You would particularly run into difficulties with your power peaks, if you would increase the power density considerably above say 10 MW

per m^3, and you would definitely have to pay penalties for reduction in your allowable power peaks.

Mr. CARLSMITH *(ORNL)* : I think the point that Mr. Jaye raises is a valid one, that one should consider all the components of the fuel fabrication cost in trying to arrive at the optimum. I think that perhaps in our paper, if we had done this, we would have changed slightly the moderator to fuel ratios and loaded the systems somewhat more heavily. I think that the effect on the fuel cycle costs would be extremely minor since the fuel cycle costs tend to be very flat with respect to change in the fuel composition.

Mr. HOSEGOOD *(Dragon Project)* : I have two questions relating to what Mr. Blomstrand said. The first one concerns the epithermal protactinium capture. I am wondering whether there is any possibility of some shielding of the protactinium resonances by any of the thorium resonances. Possibly this is not so, but I would like to know.

My second question relates to the optimisation of S-values which as I understood it, depended on the inventory costs of fissile material on the one hand versus the costs of heavy metal fabrication on the other. Now, how would this optimum be affected, if the fissile and fertile materials are separated in the feed and breed type of reactor? At first sight, the high S-value would appear to reduce the fissile inventory and fraction of power in the feed or seed zone without incurring a high penalty in heavy metal fabrication cost.

Mr. BLOMSTRAND *(Dragon Project)* : I would like to agree to Mr. Hosegood's second point. I think that one could go to better moderated systems if one separated the fissile and fertile isotopes. This means in a seed-blanket system. I am not really sure what I should say about the first question. I got the position of the resonances from Mr. Carlsmith, so maybe he should answer it. The only thing I can say is, that I do not believe there is very much shielding, though there may be some shielding of the low lying protactinium resonances by the U^{233} resonance around 1.7 eV. This is possible but I cannot really say.

Mr. CARLSMITH *(ORNL)* : I cannot recall whether there is any overlapping of the protactinium and thorium resonances. I have not looked to see whether it is an important factor.

Dr. CAPRIOGLIO : Any further comment on this problem of power density-specific power? Or S-value? Mr. Blomstrand has made another remark concerning the *leakage problem* and its importance and the incentive there would be to limit it as far as possible. Is there anybody wishing to comment on this? Everybody seems to be sharing his view, but there seems to be little to be done.

Mr. FORBES-GOWER *(CEGB)* : I do not actually want to comment directly on that point, rather more to say that one of the methods suggested is to have the

fertile material removed to the reflector. This has been in Mr. Schlösser's mind I think : to separate the thorium and to actually put it outside the core in the reflector. If after a few charges, this is generating significant amounts of power, it has to be cooled, it always had to be cooled anyway. We now have the problem that power output from this region is varying a lot during its life. This must either reflect in the higher capital cost to provide variable flow in this region on its own, either through variable gags or separate blowers, or it must reflect on capital cost through having a very poor gas outlet temperature at the beginning of reactor lifetime and the thermal efficiency is going to suffer with the blanket type system. I would like to know if in comparing the relative merits of having thorium in the core or in the blanket, such factors have been taken into account.

Mr. CAPRIOGLIO : I fear there is some sort of misunderstanding in the terminology of seed and blanket as it has been used by Dragon. It is not my undestanding that they intend to put the thorium in the reflector, but I would like them to make a comment on this.

Mr. BLOMSTRAND *(Dragon Project)* : Yes, when we speak about *seed and blanket* systems, this only refers to a separation of fissile and fertile materials so that they can be allowed different residence times in the systems, but we always assume that there is a free exchange of neutrons between these elements so that they are fairly intimately mixed. I think that Dr. Stewart already touched the point of thorium blankets in his neutron balance that he showed on the blackboard and how much that could decrease the leakage.

Mr. LOCKETT *(Dragon Project)* : As far as the heat output from the core is concerned, the channels which cool the seed and the blanket are in fact running parallel together and they mix into a common plenum chamber before entering in the heat exchangers just the same way, as if it had come from a magnox type of reactor. Does that answer the question?

Mr. FORBES-GOWER *(CEGB)* : Well, it answers it by suggesting that in fact we do suffer a degradation of the gas outlet temperature when less power is being generated in the fertile material. One thing that surprised me was, all the people talking of low neutron leakage : One of the reasons they got this was through having the fertile material in the reflector and now they say they have not put it there. How do they get low leakage while still having the fertile material adjacent to the feed material?

Mr. HOSEGOOD *(Dragon Project)* : I would like to try to attempt an answer to both questions. As Mr. Blomstrand would no doubt have told you much better than I will, the reason that we do not get a power variation from the fertile zone in the breed and feed reactor is that we prime it in the first place with fissile material. It would consist in fact of a self-sustaining mixture of fissile and fertile material, so that there is no change in its fissile content and heat output with burn-up. The seed zone is then necessary to make the reactor critical.

The other part of the question related to putting a blanket of thorium around the reactor instead of a reflector, in other words to make a complete two zone system in two widely separated regions, this would certainly incur penalties in complications. One way of dealing with the resultant cooling problems would be to use a two pass coolant flow arrangement, where the coolant flowed in series through the blanket and through the core, but this obviously complicates the engineering a good deal. Such a system would appear only to make sense really if reprocessing were definitely used to recover the fissile material from the blanket elements, so that it could be used as feed for the core.

Dr. CAPRIOGLIO : I think this clarifies the question of blanket and I think that it is why the Dragon people do call it feed and breed rather than seed and blanket, in order to make a clear distinction with the proper seed and blanket concept.

Prof. SCHULTEN *(KFA/THTR)* : I should mention that in our concept we are making investigations about the use of blanket elements which are first really blanket elements and afterwards are coming in the reactor core to be an inner blanket element. But the problem again is whether it is profitable to retain the neutrons by a thorium blanket. But when, let us say, one is trying to get a very high conversion factor, I think this could be a solution. At the same time, we believe that the problem to get the heat away is not so difficult in this case, because that means only, more or less, there is a bigger pumping power for blowing necessary.

Dr. CAPRIOGLIO : I suggest that we stop the discussion on this here and we will come back to this problem of feed and breed, because there are many other interesting questions to be discussed.

I would now go on and ask Mr. Carslmith of Oak Ridge if he has any comment on Dr. Schmidt's presentation or any further remark to give.

Mr. CARLSMITH *(ORNL)* : I would like to discuss a little further a point that has been referred too briefly already a few times and that is the question of the *assumptions regarding the fabrication and processing cost.* As Dr. Schmidt mentioned, these assumptions represent the biggest single difference among the various papers that have been given in the session. I think, we would try to distinguish between two quite different reasons for the differences in assumptions on fabrication and reprocessing cost. One is that there is a considerable uncertainty regarding the processing, the unit operations, and the total costs that are to be used; but the other and perhaps just as important, is the question of the context of industry size in which these estimates are to be made. We have tried in our paper to give estimates of the unit costs, as a function of the throughput and thus of the industry size. On fig. 5, we have shown the unit costs of refabrication and processing that we have used as a function of throughput; as one can see, we have covered a very wide range both of throughputs and consequently of unit costs. In our optimisation, we have not in general treated this item as a completely parametric factor, but have chosen a particular size of industry; the size that we have identified as being one that we wanted to look at, was a fabrication and reprocessing plant that would serve

a reactor capacity of 15,000 MW electric. This is a very large size as everyone recognises, and it is the size that we identify as being appropriate if one is thinking of a period something like 20 years from now, because at that time, if this concept is successful, one will have large complexes of plants. Now we certainly do not mean to imply that this is the only context in which it is worth looking at these reactors and it is obvious of course that one needs to also look at smaller size reactors and smaller complexes of reactors, for the purpose of deciding what the economics of the next few years are going to be. I would invite comments from the authors of the other papers as to exactly what context they were using, what period of time they were considering and what size reactors they were considering in their estimates. I think this might shed considerable light as to the reason for the differences between the assumed fabrication and reprocessing costs in the different papers. Not only does this make a difference in the total fuel cycle cost that one gets, but also makes a difference in the optimisation as one might expect. One of the points that we have found where it could make a difference, is that in looking at the optimisation of fuel management systems and fuel types as a function of unit fabrication costs, we come to the conclusion that the fully enriched uranium with thorium and with recycle is particularly attractive at low unit fabrication costs and thus might be particularly attractive at some time considerably in the future. For higher fabrication costs, we actually find a slightly lower fuel cycle cost with a partially enriched uranium fuel in which the spent fuel is discarded but not reprocessed.

Now I want to speak of one other slightly different topic on which we made some investigation, but was not covered in any detail on Dr. Schmidt's summary and that is that we attempted to look at an heterogeneous fuel element using a *low enrichment uranium*, partially enriched uranium fuel and no recycle. The object of looking at this, was to determine whether the enrichment could be reduced far enough, so that one would get a substantially cheaper fissile material than one can get with the homogeneous system. What we found was that we could reduce the enrichment by lumping the fuel as closely as possible in fuel channels and leaving large amounts of graphite outside the fuel cluster, whereas in a homogenous partially enriched uranium system we obtained an optimum enrichment of about 7 % to 8 %. In the heterogeneous case, we found that we could get down to 2 % or 3 % enrichment. On the other hand, this had to be done at the expense of reducing the power densities in the core considerably, since there is a large amount of the core then allocated to just moderator with no fuel in it. The reduction in power density and specific power appeared to at least offset any advantage that was gained due to the lower enrichment. As a matter of fact, as you recognised, the enrichment of 2 % or 3 % is still not low enough to obtain really inexpensive uranium. We were unable to get much lower enrichments than this, because in the designs we were using, where there was still no metal allowed in the core, there had to be some graphite surrounding the coolant channels and the fuel, and thus it was impossible to reduce the U^{238} resonance integral to the extent that one would like and to the extent that one can in metal clad systems, as the AGR or Magnox reactor.

Dr. CAPRIOGLIO : I think we had two main questions. One is the very important one of the size of industry and the size of total capacity, which is assumed to make the economic calculations and optimisations: the other one, is the use of slightly enriched uranium for these reactors. As a matter of fact, the second one looks to me very similar to a sort of advanced AGR, or an A^2GR.

Taking these two independently, it seems to me that this question of the size of the industry leads almost naturally to have reprocessed fuel. If one is assuming to start from the very beginning with a very large size of the industry, almost inevitably one is bound to find that reprocessing is worthwhile and that the once-through cycle is not interesting enough.

In fact, I should imagine that at short term people should be considering both cases of once-through and reprocessing, since in order to achieve such a large size, one has to go through the very painful steps of small sizes.

I am a bit surprised to see that G.A. is not considering in a very thorough way a once-through cycle, which would have the advantage of not forcing the size of the reprocessing plant to go too quickly to something which is really economic.

Dr. STEWART *(General Atomic)* : I do not think there is any disagreement with any of the things that have been said, but I think that the information we have prepared for the meeting does concentrate on the longer range, and we feel that it is essential that one gets to the position where one can reprocess fuel just as soon as possible. After all, the first generation reactors are already gaining experience, and we believe that if we are to capture a significant part of the market fairly soon, one has to look toward all features that can get the minimum fuel cycle cost as soon as possible.

Dr. CAPRIOGLIO : I think this answers my question, and this really points out a difference between the outlook that is now present in the United States with respect to the one that seems to be prevailing in Europe where we seem to be more concerned with finding out what is the minimum fuel cycle we can get with no reprocessing at all. If this is interesting enough, we think, this is perhaps the best way of getting started. Then when the size of the industry will have grown enough, then we would consider reprocessing as bonus that would be added to something that is already quite good. I am wondering if Dragon wants to comment on this?

Dr. HINTERMANN *(Dragon Project)* : I think an answer is given in the Dragon paper by Blomstrand and others, fig. 13. There the fuel costs are plotted against fabrication charges in the case of once-through cycle, or fabrication and reprocessing charges in the case of reprocessed cycles. One can see that to get the same fuel cost, in the absciss there can be a difference of about 200 £/kg heavy metal. This means, when we start with the programme of high temperature reactors, then first probably one would start on the once-through cycle, with no credit for irradiated fuel and as soon as enough unprocessed fuel was available, one could start to reprocess it with the cost of 200 pounds sterling per kilogram which is about 600

dollars per kilogram. From then on, a reprocessing would start from itself on a economical basis. With further growth of the industry, this cost will certainly diminish considerably and reprocessing will lower the fuel costs. Emphasis will then shift from high fifa to high conversion.

Dr. CAPRIOGLIO : Any further comment on this general problem of size of industry? We have been talking about the size of reprocessing facilities. Now what is the feeling of the fuel element manufacturers about the size of the fuel element production industry? Is there such a large difference as it seems to be the case for reprocessing? I have seen in Mr. Carlsmith's curves that the curve for the fuel element fabrication cost, as a function of size, is considerably less inclined than the curve for the reprocessing plants. Is this the feeling also of the other workers in this field and is there any comment from fuel element manufacturers? No? Well, everybody seems to agree with Mr. Carlsmith on this, which is very comforting by the way.

Mr. BLOMSTRAND *(Dragon Project)* : I have one comment on the low enriched fuel cycles. Mr. Carlsmith has made a very interesting comparison between the once-through cycle with low enriched fuel and with high enriched fuel on the thorium cycle and, in his table on page 363, I think, he shows a very interesting thing. I am referring to the power peak in the fresh fuel. We have generally found on the once-through cycle with uranium/thorium, that we get power peaks almost in the order of 2 in the fresh fuel, but Mr. Carlsmith is referring to a power peak in the order of 1.5 on the partly enriched uranium cycle. Now the high power peak in the uranium/thorium cycle is due to the fact that the power collapses quickly, because the uranium-233 that grows in has the same cross section as the uranium-235 that is burning out. In the case of the partially enriched cycle, the plutonium that grows in will have a higher cross section than the uranium that is burnt out. Though the fissile material is actually depleted in the fuel element, this effect will lead to a very favourable power history in the element. I want to ask Mr. Carlsmith, if he has looked into the fact of whether the initial power peak is really the highest one, or if it goes up when the plutonium in growing in and then slowly decreases. I think, that as a general comment, this partly enriched cycle is of very great interest for designers who like low power peaks.

Dr. CAPRIOGLIO : It seems to me a usual happening in these low enrichment fuel cycles to have a peak at a certain moment rather early in their life, but I would like to ask Mr. Carlsmith if this is the case,

Mr. CARLSMITH *(ORNL)* : Yes, in the partially enriched uranium fuel, the power does go up slightly from the initial value and the peak is not quite at the start of life but a little later. In the particular case that I have illustrated here, the rise is not very much and the maximum power is only a little more than the initial power. However, these power versus burn-up curves are extremely sensitive to the exact enrichment used and generally speaking, if one tries to get very long burn-ups and uses high enrichments, the power peaking rapidly becomes much worse.

Dr. SCHROEDER *(Dragon Project)* : This is a more general remark about low enrichment fuel cycles and the classical thorium/uranium fuel cycles. There is a little conflicting situation, because on the one hand we would like to build up as quickly as possible a large power industry in order to get the benefit of the reprocessing. On the other hand we would like to have attractive fuel cycles right now. The low enrichment fuel cycles seem to offer such a choice for the moment, but the conflict is that if you are going on with this type of fuel cycle, you deprive yourself of the possibility of coming to a large industry with attractive reprocessing possibilities.

Dr. CAPRIOGLIO : Any comment from Mr. Carlsmith ?

Mr. CARLSMITH *(ORNL)* : No, I think that puts the situation very clearly.

Mr. RENNIE *(Dragon Project)* : I think one should not forget that in the low enrichment case that Mr. Carlsmith quotes, the power density is appreciably lower. As I understand, it is about 2.5 MW per m^3, whereas in the comparable uranium/thorium case it is probably between 7 or 8,

Mr. CARLSMITH *(ORNL)* : The power density in the heterogeneous low enrichment system was low. In the other semi-homogeneous cases that I have described, the power density would be about the same as in the thorium system.

Mr. BLOMSTRAND *(Dragon Project)* : I have an additional comment on power densities in partly enriched uranium systems. There this protactinium poisoning does not occur, so that if one goes to higher and higher power densities and lower the fuel element residence time, this will just decrease the working capital costs without corresponding penalty in conversion ratio. Does Mr. Carlsmith agree to that ? This gives an incentive to high power densities.

Dr. CAPRIOGLIO : Mr. Carlsmith agrees. Any further comment on Mr. Carlsmith's presentation ?

I would like to go on now and ask Dr. Brandes if he has any comment to Dr. Schmidt's presentation.

Mr. BRANDES *(KFA)* : Let me make some remarks in German.

Translation

Perhaps I might describe the fuel cycle in a little greater detail. There are two types of balls. One type has thorium and uranium-233 in equilibrium. This element is reprocessed. The second element contains uranium-235 as make-up fuel and is not reprocessed.

I should like to make a further comment on the structure of the higher uranium isotopes. It is certainly not correct that the chain stops at uranium-234. I have therefore recently investigated a case not described in this paper in which it was assumed that uranium-234 and the uranium-235 which is in the process of formation are present in equilibrium concentration. For the uranium-236, which is also in the process of formation, a build-up time of 15 years was assumed. In this way, the

neutron absorption in uranium-234 is practically replaced by neutron absorption in uranium-236 and in the fission products of the newly formed uranium-235. The calculations showed no deterioration of the optimum physical conditions.

As far as the fuel costs are concerned, I should like to add that in view of the wide divergence of opinion concerning the individual costs, the figure of 0.65 mills per kWh should be understood rather in a relative sense. The influence of the various parameters on the minimum fuel cycle costs should be shown. The cost calculation is therefore based on very simplifying assumptions.

SCHLOESSER *(Euratom/THTR)* : The equations on page 374 of your paper relate to the make-up fuel for U^{235} in a fuel cycle whereby thorium, protactinium and U^{233} are in equilibrium. The higher isotopes are, however, not mentioned in the equations. Is it possible that these were in fact considered in the calculations, but the equations are not presented in that form? Perhaps it is merely a misunderstanding.

Mr. BRANDES *(KFA)* : What you say is quite correct, but I have in addition carried out some calculations, not published here, in which uranium-234 and uranium-235 are in equilibrium and in which assumptions were adopted also for uranium-236. The calculations showed that the deviations were very small.

Dr. CAPRIOGLIO : I would like to ask now Mr. Giesser, if he has any further comment on Dr. Schmidt's presentation.

Mr. GIESSER *(BBK/THTR)* : Only a few remarks are necessary, I think. A first remark is concerned with the *fuel cycle in the THTR*. The fuel elements are continuously supplied and withdrawn and are circulated with a high recirculation rate. In this way, no excess reactivity has to be built in, perhaps an advantage in comparison to the other reactor types. Our calculations are performed for an equilibrium state independent of time. In this model the average of the concentration vector in the core is calculated by the time average. We have assumed that all fission products are completely retained in the fuel elements. A total neutron leakage of 5 % has been considered throughout. As pointed out by Dr. Schmidt in his review, the difficulties arising in connection with an unsaturating build-up of U^{236} are avoided by proper fuel management. We have assumed 2 or 3 types of coated particles differing in size. A corresponding initial condition for the concentration vector may be formulated. Perhaps Dr. Stewart and his co-workers will give further comments on this point.

Another remark is concerned with the assumptions made in our cost estimates. In the present calculations the costs for the first core are assumed to be capital costs, which are not considered here. As indicated by Dr. Schmidt the fuel cycles costs of Dragon and BBK differ by 0.2-0.3 mills/kWh. This difference is due to the interest and taxes paid for the first core fuel inventory. I agree that this cost item should be included for reasons of comparison.

The calculation of the fabrication costs for the first core depends to some extent on what financing procedure is applied. This procedure may vary from country

to country, so that difficulties arise if one wishes to standardize cost calculations. A final remark is to be made on reprocessing costs. These costs are small in comparison with the fabrication cost and the cost for fissile material. This is to be expected because of the relatively high burn-up achieved. Therefore we have a relatively small throughput of heavy materials in the processing plant.

Dr. CAPRIOGLIO : I would like to take first this question of *putting the investment charges into the capital cost of the plant rather than into the fuel cycle.* I feel myself, that this is an unwise procedure, because when one wants to make a general optimisation one wants to keep the fuel cycle separate from the capital cost.

Although there are feedbacks, as we have found already this morning, one would like to keep these feedbacks as small as possible in order to keep the ideas as clear as possible.

Putting the first charge into the investment, is a rather confusing method as far as I can judge myself, because one is always sort of coming back and forth between the capital cost and the fuel cycle cost. I should like to invite opinions on this.

Dr. STEWART *(General Atomic)* : I simply want to say that I agree 100 %.

Mr. BLOMSTRAND *(Dragon Project)* : The results will be then dependent on the assumed reactor lifetime, because the influence of these initial costs, if one puts them in the fuel cycle costs, will depend on how long one considers the reactor to run, if one wants to amortize them over the reactor lifetime. I think, if one wants to make comparative cost studies, one can do it either way, just to see what happens if one is comparing different systems.

Dr. CAPRIOGLIO : This introduces a new element of doubt in our minds. I think myself that Blomstrand's remark is very valid, but not enough to shift my opinion. What about Dr. Stewart?

Dr. STEWART *(General Atomic)* : I am still with you 100 %.

Dr. SCHMIDT *(BBK/THTR)* : I think there are some doubts on the fabrication costs, whether interest should be paid on this one. But it would certainly be much easier to compare different calculations, if at least the interest in the fissile material would we included.

Dr. CAPRIOGLIO : The question of U^{236} *build-up* has been raised and I think Dr. Stewart is likely to make a contribution; therefore, I would like to ask him once again to speak.

Dr. STEWART *(General Atomic)* : We have given a lot of attention to various fuel management programmes in our work. I think that the importance of the fuel management was first pointed out by Mr. Jaye, so far as I know several years ago, and I have here some pictures which maybe will illustrate the differences between the various possibilities (See fig. following pages).

What we have shown here, is two successive fuel cycles. That is, the block here indicates the fuel element in one cycle, the next block the fuel element in a succeeding cycle. Now, this particular schematic refers to what we call a *full uranium recycle management programme.* Here we take the uranium which consists of U^{233}, U^{234}, 5 and 6 from the previous cycle and feed it into the fuel element in cycle N. In addition we put thorium into the fuel element and we put in some U^{235} which is make-up fuel in order to compensate for the difference in conversion ratio between unity and something less than unity. At the end of this cycle, we remove all the bred fuel, simply relocate it in the new fuel elements together with the thorium, add some make-up U^{235} and some make-up thorium. This cycle is then N plus 1. So you see what you have done is to carry along completely all the uranium isotopes that are built up plus to add a significant amount of U^{235} which builds up U^{236} and neptunium quite quickly.

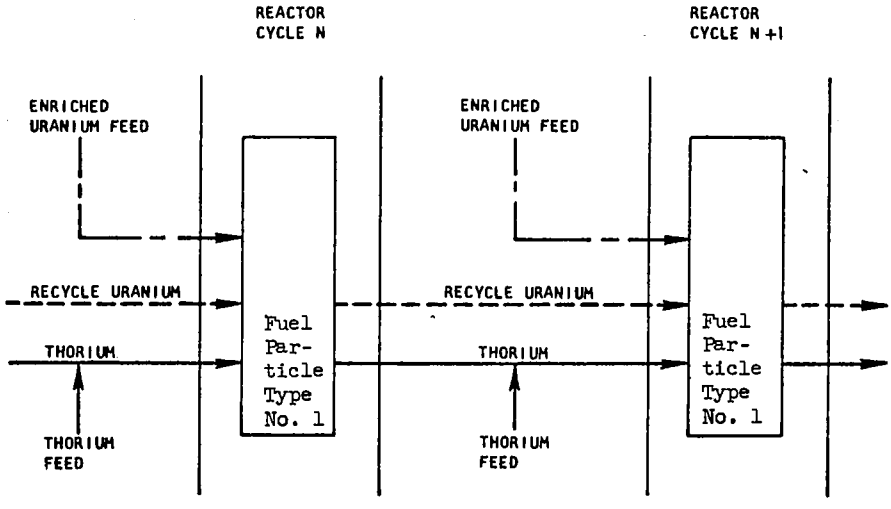


FIG. 1. — Schematic of full uranium recycle fuel management programme

Now, we can go to the next one. This is what we refer to as a *once-through uranium recycle programme* and this is to be distinguished from the use of the word once-through as it has been previously used today. The thorium particles are coated separately from the fuel. At the end of the cycle, we remove the U^{233} which has been bred in the thorium and recycle it together with make-up U^{235} in a separate particle. At the end of the cycle then, we remove the U^{235} and U^{233} which is left over; this may amount to something like 10 % of the fuel altogether in the fuel element at this time. This fuel residue is then shipped off to some other reactor such as a burner reactor of some kind and used in some other way. We then take just the U^{233} that is available from the thorium particles and again replenish it with U^{235} and move on to the next cycle. The advantage of this type of fuel management is that we limit the amount of U^{236} which is built up only to the U^{236} from the U^{235}

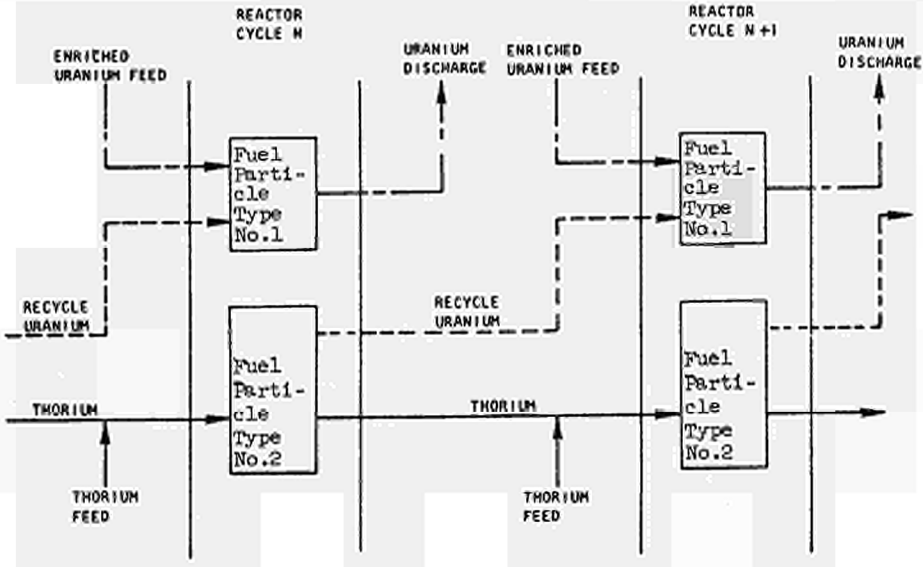


FIG. 2. — Schematic of once through uranium recycle fuel management programme

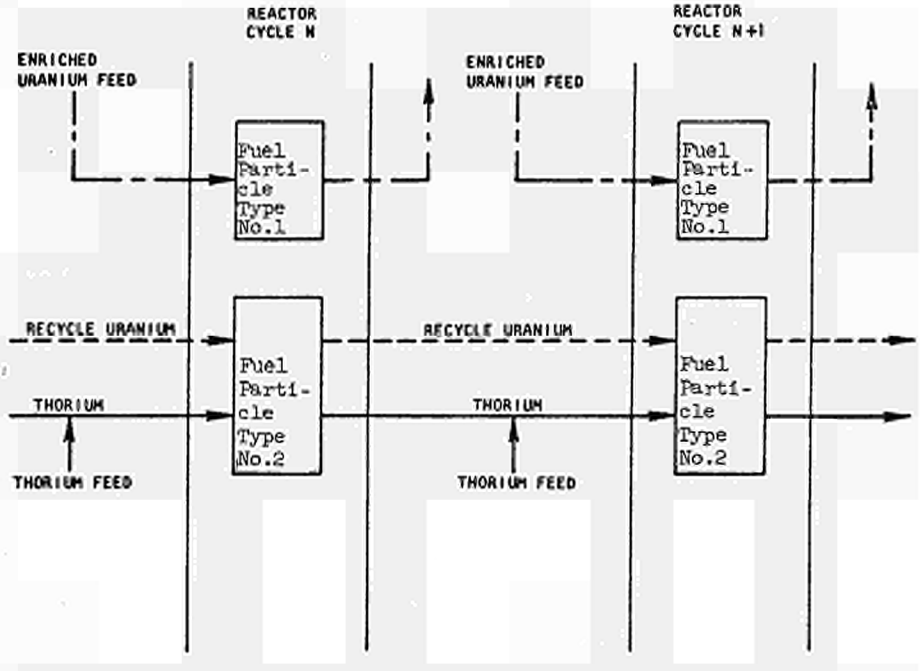


FIG. 3. — Schematic of bred uranium recycle fuel management programme

feed, and in addition we cut off the build-up of U^{235} which is not as good a fuel as U^{233}.

Now the next fuel cycle management programme is what we call the *bred uranium recycle fuel management programme.* Here we coat the thorium plus the U^{233} fuel that has come from the previous cycle in one particle and coat the U^{235} make-up fuel in another particle. At the end of this cycle then, we take the U^{233} out that has been bred, plus any of the higher isotopes that have built up in the meantime and again coat that as a separate particle, again keeping the U^{235} feed separate from the U^{233} and thorium. In this case at the end of the cycle, the amount of fuel that we export, as we call it, is only about 5 % of the total fuel that is in the fuel elements, so that one does not have to export quite so much fuel in this case. On the other hand, it has the disadvantage that a little bit of U^{234}, U^{235} and U^{236} will ultimately build up in the recycled uranium. Of course, the amount of U^{235} that builds up in the equilibrium case is just equal to the value of alpha for the U^{233}, so that in the equilibrium, one gets something like 10 % U^{235} relative to the 90 % U^{233}, but in addition of course there is some U^{235} from the feed, depending on what the conversion ratio is.

Now, just let me say a word or two about the relative conversion ratios that one might get under these various management programmes. We find that the conversion ratio is very, very sensitive to the fuel management programme, if one has conversion ratios in the neighbourhood of say 0.8 or lower. If it is below 0.8, then we would find that the conversion ratio deficiency due to the complete recycle might be something like 0.10 or even larger, relative to the two fuel management programmes I have talked about subsequently here. The difference between the bred uranium recycle and the once-through uranium recycle is about 0.01 or 0.02, so that it is relatively small and does favour of course the once-through uranium recycle. On the other hand, remember that one exports a larger amount of fissile material in this particular programme. After one gets to conversion ratios of above 0.90, the differences get considerably smaller. The conversion ratio for the complete recycle is smaller perhaps by 0.05 relative to the various other programme schemes, so that if one can get good conversion ratios to start with and therefore not require very much uranium feed, that is U^{235} feed, then the specialized fuel management programmes are not quite so important. In conclusion let me just apologise to Mr. Jaye for talking about a subject that is near and dear to his heart.

Mr. JAYE *(General Atomic)* : That's all right.

Dr. CAPRIOGLIO : I think we all an impressed by the sophistication of this work. Is there any comment ?

Mr. CARLSMITH *(ORNL)* : Let me just make a brief comment which is really an extension of what Dr Stewart was saying, and that is that if one is using full recycle of all the fuel and the conversion ratios become very low, the situation becomes extremely serious in that below a conversion ratio of about 0.7, the recycle material can actually become more of a poison than help to the fuel system and it

is not worthwhile to recycle it even if the reprocessing costs are zero. It had better be thrown away.

Mr. JAYE *(General Atomic)* : I might make just one very, very short comment, that it is pretty difficult to get the conversion ratio above 0.90 if you do not have one of these fuel cycling schemes, and so while on the one hand it gets to be less important, on the other hand it gets to be very hard to achieve.

Dr. SCHLOESSER *(Euratom/THTR)* : I would like to ask Dr. Stewart about the exported fuel. How is the exported fuel accounted for in the fuel cost analysis? I mean, is it completely discarded, as for instance in the once-through fuel cycles at Dragon, or is there any credit given and how has this been calculated?

Dr. STEWART *(General Atomic)* : In our calculations, we have given it full credit, namely 14 dollars per gramme. This is perhaps unrealistic, as has been pointed out by the Oak Ridge people. They assign a value of something like, I believe, half the normal value of U^{233} to the exported fuel. We feel somewhat justified in using the full value for the exported fuel, first of all because it is not a terribly large effect though it is significant, but secondly because we feel that the value of the U^{233} is probably somewhat higher than 14 dollars for the material that has been recycled in the fuel management program.

Mr. DE BACCI (*Euratom*) : I want to raise a question about this export fuel I hope by the way they do not mean it literally. When examining the implications on the conversion factors, especially if one looks at a self-sustaining breeder or quasi breeder economy, this exported fuel is in a way lost. Although it is still fissile, it is not fissile that you can use within your system and if you want to sustain your quasi breeder industry, you have to mine a new fissile, is that not so?

Dr. STEWART *(General Atomic)* : That is so but I want to make some further comments on the importance of conversion ratio when you call on me to discuss my paper.

Dr. CAPRIOGLIO : I would like to ask now Prof. Schulten to comment on the *breeding in pebble bed reactors.*

Prof. SCHULTEN *(KFA/THTR)* : We have tried to find out the conditions which are necessary to come from the high conversion factor to a real breeder factor. This consideration is not connected with any economic question; it is only a study to point out a way to get a real breeder factor. Now, I think the easiest way to clarify this is perhaps to make a comparison between the results which are given by Dr. Stewart and in our paper. (See table).

When we consider the losses, first the leakage, Dr. Stewart gives in his third case a leakage of 0.05, and in our case, where we included a blanket, we got a leakage of 0.02. For the moderator losses the first case is 0.03; we got losses of 0.05, because we have a higher moderation ratio. For the protactinium, U^{236} and xenon losses in both cases we got the same result.

	General Atomic	Schulten	Balance
Losses :			
Leakage	0.05	0.02	+0.03
Moderator	0.03	0.05	—0.02
Pa	0.03	0.03	—
U^{236}	0.01	0.01	—
Xe^{135}	0.04	0.04	—
Fission products	0.10	0.03	+0.07
Control	0.03	—	+0.03
Eta-value	2.16	2.22	+0.06
			+0.17

In the case of the fission products, I should explain a little more. In our fuel management, as mentioned before, we are thinking about two types of elements, the one which contains only fuel and the other one only thorium. The thorium elements are first used as an outer blanket and afterwards as an inner blanket. So the concentration of fuel is built up in the normal way. The residence time of the seed elements is much shorter than the residence time of the thorium elements, so that the input for the reprocessing plant is rather small (measured by kilograms of fuel), because we have to reprocess very often the fuel itself, but relatively seldom the thorium. This different residence time gives a gain of neutrons. When you reach a burn-up of about 30,000 MW days per ton, in our case the fission products should be in the order of 0.03 and in the first case it had been 0.10.

The next item is the control; we did not consider the control losses.

The next point is the eta-value which is one of the most important factors. In the calculation of Dr. Stewart, we got 2.16, in our case we got 2.22. There are two reasons that I can see for the difference : the first is that the General Atomic case has much more U^{235}, making the eta-value smaller; in our case the moderation ratio is higher so that the spectrum is softer and this contributes also a gain in the eta-value.

Now, when we take the gains and the losses together with the difference in the eta-values, the result is a net gain of 0.17. When we take the conversion factor which is calculated by Dr. Stewart, we have to add 0.17 to 0.87, that is 1.04, representing the difference in the calculations.

We have also calculated the case discussed by Dr. Stewart with our computation and we are in relative good agreement except for fission products. Perhaps we have too optimistic values, but this is more or less connected with the data for fission products, which are not quite clear. This influence in the data for fission products is not so important when the residence time of the fuel is very short because then the contribution of these errors is rather small.

Now, I think from a technical standpoint, it is necessary to develop a system where a short time fuel handling is possible. My opinion is that this could be made only when loose coated particles are used and when the fuel management is made in a continuous way. I should mention finally that in our calculations, there might be some errors because they are preliminary calculations. I am quite convinced, however, that when beryllium oxide is used as a spine of fuel elements, and when it is possible to solve the problem of lithium 6 (perhaps solved by one of the several methods to remove it, such as by a by-pass heat treatment or something like this), then I am really quite convinced that the development of a high temperature reactor, not in the near future but in the far future, can lead to a real breeder reactor.

Mr. JAYE *(General Atomic)* : I would like to ask Prof. Schulten two questions. First, I would like to know what the fifa is in this system because indeed the fission products are very, very low. We normally calculate about 0.10 for about 1 fifa; I think this must be 0.2-0.3 of a fifa.

Prof. SCHULTEN *(KFA/THTR)* : I think it is not so easy to speak about fifas because it is quite another fuel management. A quite different definition of fifa is necessary in this case; most of the fuel is bred in the thorium, it must be transferred to the fuel particles and the fifa of the fuel particles itself can be rather high.

Mr. JAYE *(General Atomic)* : Perhaps I do not really appreciate this fuel management scheme in total and this may be my main difficulty. However, I think if you have a large fifa as averaged over the entire core, then it seems to me this is a direct measure of the fission product losses; I am wondering what this fifa is.

Prof. SCHULTEN *(KFA/THTR)* : I think it is rather difficult to make a definition of an average fifa in this case. I must say that the fifa in this case is relatively low; lower than 0.5. There is some gain by dividing the fertile material and the fuel material in two parts, where the fuel material is reprocessed very often and most of the fissions are made in the fuel particle, not in the thorium particle. This makes a big difference compared with a normal fuel scheme where you have together the fuel and the thorium.

Dr. STEWART *(General Atomic)* : I think we generally agree that the trends are in the direction that you mentioned here and we hope you are right. We would point out that on the fission product poisoning, if you look at the poison as a function of fifa for example, you find that it builds up quite rapidly, as you know, early in life : then it assumes some sort of a linear variation after the first 0.1 or 0.2 fifa or something like that, so that we would be a little surprised that it would drop off all the way from 0.10 to 0.03, by reducing the fifa say from 1.0 down to 0.5 or even 0.4. As I say, I hope you are right on this. The other differences, for example, by cutting the leakage down by surrounding the core with a blanket, could of course be used in other types of high temperature reactors. We have not considered it for the reasons pointed out by Dr. Sanders a while ago, namely that it does create some rather difficult power peaking problems, or I should say, degradation of the tempe-

rature of the coolant. I only showed it in my one slide to show what one could get, if you considered everything that might be available to you. I am going to say something about conversion ratios later on.

Mr. CARLSMITH *(ORNL)* : I have a suggestion for those designers that present designs which lead to breeding in this type of reactor system and that is that I think it would be wise for them to make an effort to calculate fuel cycle costs to go along with the presentation. We recognise of course, that it is difficult sometimes to agree on unit cost to be used. We are also looking at breeders for some time in the far future, but on the other hand it will still be necessary that a reactor operates in an economical fashion under some set of ground rules. Breeder designs normally take advantage of more exotic materials such as BeO or very short lifetimes or blankets. Particularly in the case of blankets, it is often surprising how much this can add to the fuel cycle cost, when one actually tries to calculate what it requires to get a thick enough blanket to really reduce the leakage significantly.

Dr. SCHLOESSER *(Euratom/THTR)* : Although I agree basically with Mr. Carlsmith I think it might be interesting to see how far one can push this system and how high the conversion factor can be made, if one makes some admittedly optimistic assumptions. It is just to show how much one has achieved with these more conventional types of reactors. I have prepared a slide; this may also clarify somewhat the fifa question (the slide is reported in page 518).

The slide shows the overall conversion factor versus burn-up for a large pebble bed reactor. The X-axis is fifa and on the Y-axis you have the overall conversion factor. The three lines give different moderator ratios, this means graphite to fissile uranium ratios. There is an optimum around 5,000 which is not very surprising when one looks at the eta-value for U^{233} dependent on the spectrum. It can be seen that for fifa about one, one gets a conversion factor of one and that for fifa below one, one gets a certain breeding. Now there are two effects, I would like to mention :

— The leakage is 1 % but this is not to be compared with the figure which was just mentioned, because it is 1 % of the neutrons absorbed. Neutron balances shown before are concerned with neutrons produced.

Now the buckling for this leakage is just about the same size as in Mr. Carlsmith's paper. The leakage is reduced because these systems are more heavily loaded than Mr. Carlsmith's systems which are calculated with moderator ratios around 10,000 and above.

— The effect which makes a conversion factor above one for short-burn-ups possible is that short burn-ups and a conversion factor above one, one can take the uranium out of the system. Then the uranium isotopes saturate and the equilibrium between the different uranium isotopes changes in such a way that one obtains more and more absorptions in U^{233} and less absorptions in U^{234}, U^{235} and U^{236}. This means that when decreasing the fifa value, the eta value of all the uranium isotopes increases because of the removing of uranium isotopes out of the reactor.

This fuel cycle is in equilibrium and it has to be started off with segregated fuel, as otherwise too much U^{236} or let us say U^{235} and U^{236} will be in the system, so that one does not get over a conversion factor of one, roughly speaking. It is only when one uses segregated fuel that right from the beginning one has a high eta-value so that one can run this system until it comes into equilibrium and these figures refer to the equilibrium cycle.

Dr. SCHROEDER *(Dragon Project)* : I would like to ask a question to Prof. Schulten and I am referring to fig. no. 1 in his report. If I understood this figure well, he has seed material lifetimes as a parameter and his abscissa represents lifetime of a breed sphere. I am astonished that he gets a minimum in the total poisoning after 1 or 2 years; one would suggest that this poisoning is less the shorter the exposure times for the blanket or breed. Therefore, one would expect these functions to rise right from the beginning and not to show up a minimum.

Prof. SCHULTEN *(KFA/THTR)* : We have also been very surprised about this fact. I can say that we have had some days of a very intensive discussion about this effect; but this effect is real and I will try to explain it very shortly. The point is : when you have fixed the lifetime of the seed element and you are going to shorter and shorter residence times of the blanket element, then more and more fuel is burnt in the seed elements. The burning of fissile material in the seed elements has a disadvantage compared with burning in the thorium elements, because there we have to pay more for the fission poisoning of the seed elements than for the fission poisoning in the thorium elements. The build-up of the fission products is different in both types of elements as it is shown in figure 2 and 3 of our paper I think, this is the reason. We are quite convinced that this effect exists.

Mr. CAPRIOGLIO : I would like to invite more comments on this question of breeding, although I would like to somewhat anticipate what we are going to talk about further on, saying that I do not really see why we attribute so much importance on this sort of discontinuity of breeding ratios of 1. It seems to me that Mr. Carlsmith's remark about how much it costs to achieve something is the real key question.

Prof. SCHULTEN *(THTR)* : I should make a very short remark to Mr. Carlsmith's remark. I think he is right; we also are only trying to build reactors which can be sold, but on the other side I think it is important to know the conditions which are really necessary to get breeders. I think that is a very important question and should be investigated. We should have the conditions ready for fuel manufacturers and for reprocessing experts, to say what we really need. Then we should see in the next 10 or 20 years whether they can go on step by step and nearer to high conversion factors. I believe that the figure for conversion factor is a function of the technology of our fuel.

Mr. HOSEGOOD *(Dragon Project)* : I would like to ask Prof. Schulten if he can give us some of the size data on the type of system he has analysed. We have

heard that it has a high S-value and we have heard that it has a high heavy metal loading; we know that the pebble bed has a high voidage and we have here a two zone system with a very low leakage. These appear to indicate to me an extremely large reactor in terms of volume. I would like to know what the dimensions are and what the power density is. I must apologise for having been unable to find them in the paper which I perhaps have not read as thoroughly as I should have.

Prof. SCHULTEN *(KFA/THTR)* : We are considering the power density in the order of 5 MW per m^3. The reactor will be a very big plant with a minimum of 1,000 MW. The diameter of the reactor is not so important because there will be a blanket which will take out the neutrons which are coming out of the reactor and there must be a minimum height of, say, 7 or 8 m. The fuel content per ball is depending on the thorium cross section and will be in the order of 0.4 or 0.5 grams per fuel element and, as I remember, the thorium content will be in the order of 12 to 15 grams per breeding element.

Dr. SANDERS *(UKAEA Winfrith)* : We have heard of several schemes using segregated fuel in which the fertile and fissile materials are initially in different fuel particles or different fuel elements. I wonder whether this introduces any problems on the temperature coefficient of the reactors. Would anyone like to comment on this problem?

Dr. CAPRIOGLIO : This is a general problem of segregated fuel which is not directly related to our breeding problem for the moment, but I invite comments.

Dr. STEWART *(General Atomic)* : I just wanted to ask another question to Prof. Schulten. When you speak of power density, do you include the blanket in the core or is this a power density just for the fuel region of the core? If it is not included in the core what would the effective power density be when you include the blanket?

Prof. SCHULTEN *(KFA/THTR)* : This is the power density per m^3 volume, excluding the outer blanket.

Dr. CAPRIOGLIO : Can we take up Mr. Sanders' question about the *influence on temperature coefficient on segregated fuel*?

Dr. STEWART *(General Atomic)* : Yes, we have given some thought to this. There is going to be some effect of course, but it depends a little bit on how one uses the segregated fuel. In the concepts we have considered, we would plan to put the fuel particles and the thorium particles in the same compact so that the delay time for the temperature to extend from the fuel to the thorium would be fairly small and therefore the thorium would respond, we think, fairly fast to the changes in fuel temperature. In addition, of course, one would program the fuel management in a way such that the thorium would normally have quite a bit of fuel mixed in it after it had been in the core for a while; so that it is only at the first part of the reactor life that this might be a problem.

Dr. CAPRIOGLIO : I would now like to ask Mr. Wagemann to make his presentation or comments on Dr. Schmidt's summary.

Mr. WAGEMANN *(KFA)* : Yes, I think I should add something to my paper. Independent from the method applied in this paper, it is possible to find out a condition by a simplified consideration which must be fulfilled in order to cope with the *available low cost reserves.* These considerations which also show the great influence of the specific power refer to a date at which already a breeder generation is installed, for instance 1980, 1990 or 2000. The fissile material which is consumed prior to that date remains unconsidered. Most authors who have made prognosis of the future demand of nuclear energy are in agreement that there is a nearly linear increase of nuclear energy demand which can be written as

$$P(t^*) = b \cdot t^* \qquad (t^* = t - 1980)$$

$$\text{with} \quad b = 71 \cdot 10^3 \ \frac{\text{MWe}}{\text{year}}$$

From that date the amount which must be mined will be

$$M = C \cdot P(t^*) - \int_0^{t^*} \frac{CP(t^*)}{Td}\, dt^*$$

with Td = doubling time

C = specific inventory $\frac{\text{kg}}{\text{MWe}}$

By differentiating we get the date from which no fissile material needs to be mined

$$\overset{\circ}{M} = O = C \cdot b - \frac{C \cdot b \cdot t^*}{Td}$$

This equation gives $t^* = Td$

By combinating this value with the first equation we find the amount to be mined to cover the energy requirements

$$M = \frac{1}{2} \cdot b \cdot (CTd)$$

Another possibility is to regard the fuel reserves and to find out the necessary product of the specific inventory and doubling time, to have sufficient reserves

$$CTd = \frac{Mv}{35.5 \cdot 10^3}$$

with Mv = available amount of fission material (kg).

The following table shows the product of specific inventory and doubling time (in America the socalled "figure of merit") for various estimates of reserves of fissile material, the sources of which are discussed in the paper.

Source	US Reserves (kg U/)	CT*d*
A	$1.49.10^6$	41.97
B	$2.87.10^6$	80.84
C	$6.74.10^6$	189.85
D	$13.96.10^6$	393.23

If these values for the "figure of merit" cannot be kept in a futural breeder generation, it certainly could be predicted that low cost reserves are not sufficient to cover the nuclear energy requirements considered here.

As mentioned before, the reserves consumed in converters were not considered; it is therefore necessary not to come up to the values mentioned above.

From these considerations as well as from the total report one cannot draw inferences about questions referring to costs and economy.

CAPRIOGLIO : I would suggest that we ask now Dr. Stewart to make his presentation and I am sure all of us will remember what he said by this afternoon.

Dr. STEWART *(General Atomic)* : I would like to add a few comments to what already has been said about our paper as reported by the rapporteur. In particular I would like to emphasize a few points we tried to make and try to put the whole question of *resource utilization and economics* into what we consider to be a proper perspective. I would like then to address my remarks to two different areas; first the importance we attach to conversion ratio, and second, the importance that we attach to low fuel cycle costs. If one is not careful, one can attach, we believe, too much attention to conversion ratios, particularly conversion ratios in excess of unity. So in our paper, we have tried to make two points that we think are quite important with regard to the utilization of resources.

First we have tried to emphasize that conversion ratio itself is not an important factor in minimising the uranium that one has to mine to sustain a nuclear business.

The second point is that the conservation of uranium in itself is also not important in a long range utilization of nuclear resources. Unless people are familiar with these arguments, these may come as startling statements. But I want to illustrate why we feel this way. To do this, I would like to project a few of the figures that appeared in our report.

The first slide indicates the uranium requirements that might be expected if we continue to use what I would refer to as first generation reactors (Fig. 3.1) These reactors typically have thermal efficiencies in the neighbourhood of 32 %, systems specific powers of around 800 kW per kilogram, based on the fuel both in the reactor and that tied up in the reprocessing and fabricating plants, and finally conversion ratios in the neighbourhood of 0.6. This is typical of the low enrichment uranium reactors that are being used today. The curves indicate the fuel requirements assuming that the bred fuel, plutonium in this case, is actually recycled through the reactor. If

one does not take this into consideration, the requirements are even larger than shown. On the left side of the figure, we have shown the amount of the uranium resources that are estimated to be available in the United States. Incidentally these figures apply only to the United States, but similar sorts of arguments apply to the rest of the world. The bars on the left side of the figure indicate the amount of the uranium that can be mined for 5 to 10 dollars, that from 10 to 30 and that from 30 to 50 which extends beyond the top of the figure. These numbers are numbers that are commonly quoted by the AEC in our country. You see that there are two different kinds of requirements for fuel :

— there is a requirement for inventory as mentioned by the previous speaker which simply is the fuel required to start up new reactors;
— then, in addition, there are the fuel requirements for burnups assuming that the reactor has a conversion ratio less than unity. In this case, you see that the low cost uranium resources would be exhausted by sometime around 1990 or 1995.

Now I would like to pass on to the next figure to show the conditions with an HTGR type of reactor, and here I want to point out several important points (Fig. 3.2) First of all, the inventory requirements are considerably lower, both because of the higher thermal efficiency and the higher system specific power. In addition the burn-up requirements are considerably lower, but let me point out that we have shown two curves here, one for conversion ratio of 0.90 and the other for 0.95. The point that I want to emphasize is that the depletion requirements are small relative to the inventory requirements, and this brings me to the first conclusion that I stated before I started this discussion, namely, the conversion ratio in itself is not an important factor in conserving resources. We have looked at the same kind of characteristics for fast breeder reactors. As you probably realize, fast breeder reactors tend to require larger inventories than the HTGR type or system so that one generally finds that the inventory requirements are larger, though, of course, the conversion or depletion requirements are negative in this case. But we find that unless one can get conversion ratios in the neighbourhood of 1.3 to 1.5 or higher, one really does not improve very much over an HTGR system. Therefore, we conclude that independent of the type of reactor that one goes to, one is probably going to exhaust the low cost uranium resources at sometime around the year 2,010 or shortly thereafter and possibly even before. So this leads me then to the second conclusion that I stated earlier, namely, that the conservation of resources is not important in itself. We are going to run out of low cost resources almost independent of the kind of reactor systems that we will install in the next few decades. Therefore the important thing, we believe, is to look at the fuel cycle costs, when one has to use the more expensive uranium ores. On this, we have put most of our attention in our subsequent analysis. So, therefore let me again re-emphasize the main conclusion of this : namely that the important thing in the utilization of all our nuclear resources is that we must be able to use economically the uranium ores that cost more than 10 dollars per pound. When we say maximum utilization of resources I want to emphasize the word "utilization" and not conservation.

Now, if I can go to the next slide, Table 4.2 I have laid out here the assumptions that we have used in calculating our fuel cycle costs. We have assumed a separative cost of 30 dollars per kilogram which might actually come down somewhat as the technology in separating U^{235} improves. We have assumed a U^{233} to U^{235} value ratio of 14 to 12. There is often the argument that the plutonium value will rise as fast reactors become more important. But we find that actually the fuel cycle cost is not greatly affected by these ratios. In the heavy water reactors, we have assumed a heavy water value of 20 dollars per pound or 44 dollars per kilogram. It may be somewhat less than this, but I think you will see later that we have been rather generous in estimating fabrication costs and reprocessing costs for this reactor, so that I do not think we have overly penalised the heavy water reactor in terms of what we might expect for heavy water cost in the future. We have taken the working capital interest rate at 10 % and fuel turn around time at 1 year. Now I have listed independently the characteristics we have assumed for the light water reactor, the heavy water reactor and the HTGR. Consistent with Mr. Carlsmith's earlier comments, I would like to point out that we have assumed in all of our calculations that this applies to a nuclear industry something like 10 to 20 years from now, so that we are assuming that there is a large production rate of fuel and a large rate of reprocessing. We therefore would anticipate that the fabrication cost for the light water reactor would come down to as low as 50 dollars per kilogram. For the heavy water reactor we have assumed even more spectacular improvements (and we may have been overly generous in this). For the HTGR we have also assumed the rather optimistic fabrication costs of 100 dollars per kilogram. Furthermore we have assigned a cost of 6 dollars per kilogram to the finished graphite in addition to the 100 dollars per kilogram associated with the coating of the fuel. We have taken up the reprocessing costs at 30, 20 and 50 for the three systems. The fuel burn-up times are 4 years for the light water reactor, 1 for the heavy water reactor and 4 for the HTGR. I do not think that it is particularly important for me to go to the other characteristics; if you are interested in studying them, you can look them up in the report.

The next slide (Fig. 4.1) indicates how the fuel cycle costs for these various reactors might appear as a function of the ore cost supplied to the reactors. Again let me emphasize that these calculations are based on technology, we would expect in 10, 15 or 20 years from now. If one would like to look at fuel cycle costs as they appear nowadays for the LWR and the HWR, I would refer you to the paper by Vallance that was presented at the Geneva conference. He indicates fuel cycle costs of around 2 mills/kWh for both the heavy water reactor and the light water reactor on the basis of today's or near term future technology.

Let me also point out that for the heavy water reactor, I have included in the fuel cycle cost, the working capital cost assigned to the heavy water. This amounts to something like 0.5 mills/kWh, but I think properly should be included as part of the fuel cycle cost, simply because this is a cost beyond that you would have either in the LWR or the HTGR. Now the dashed line refers to a critical cost that

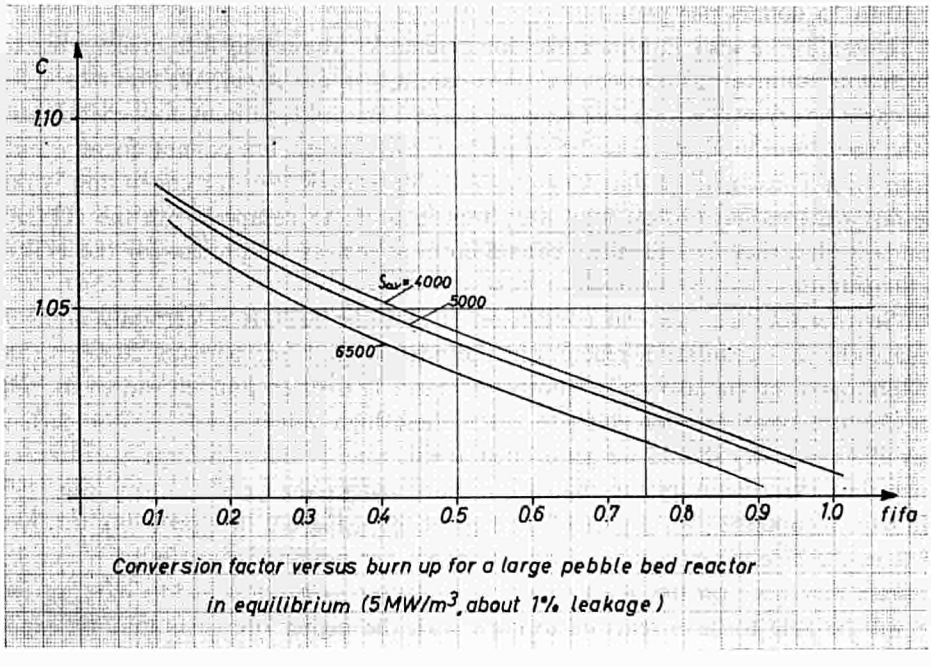
we believe has to be met in order to compete with coal or fossil fuels in the United States, taking into consideration that the capital cost of a fossil fuel fired plant will be something like 20 dollars per installed kilowatt, less than that one might achieve with a nuclear plant. Taking typical or average coal costs for the country, which turn out to be something like 23 cents per million BTU and a typical efficiency of around 38 % for coal fired plants, the critical cost, when one subtracts off the amount assigned to the extra capital cost appears to be around 1.7 mills/kWh. So this is the fuel cycle cost one has to be able to achieve, in order to compete over a long period of time with fossil fired power plants. You can see here that the HTGR does quite well, even for ore costs as high as 40 dollars per pound. The LWR and the HWR, on the other hand, exceed the critical cost for ore costs in the neighbourhood of 10 to 15 dollars per pound.

I would like also just to make some remarks about the fast breeder reactor which you can investigate somewhat more carefully in our paper. We find that unless one can do very well with a fast reactor, again I am talking about conversion ratios in the neighbourhood of 1.3 to 1.5 or perhaps even larger, one cannot do very much better with a fast reactor than one can with the HTGR. So our conclusion is that the fast reactors have to do very, very well in order to compete with the HTGRs, assuming that they can get their capital costs as low as we project for the HTGR in the future.

So therefore our general conclusion is that the HTGR is certainly going to replace the first generation reactors in the next 10 to 15 or perhaps 20 years, and on this point let me also make another remark. It is often pointed out to us "Oh, but the ore costs are not going to reach these high values until 1990 or perhaps year 2000 and why should we get excited at this time ?" But it is important to recognize that the reactor plants that are constructed in the year 1980 or even 1975 will have to operate over a part of their plant life using ore that probably will have costs in the neighbourhood of 10 to 20 dollars per pound. So we feel that one has to press very hard on the development of advanced converters at this date, so that we will be able to have reactors to cope with the higher ore costs that we expect in the next 30 years from now.

Returning then to my summary, we feel that the advanced converter such as the HTGR will definitely replace the first generation reactors in the next 20 years from now. We also feel that the fast breeder reactors probably will not replace the advanced converter reactors for some time to come, until they can develop their technology to the stage where they can ensure conversion ratios well above 1.3 and fairly large specific powers.

Dr. CAPRIOGLIO : I would suggest, we adjourn our meeting till this afternoon.



Conversion factor versus burn up for a large pebble bed reactor in equilibrium (5 MW/m^3, about 1% leakage)

PLUTONIUM FUEL CYCLE STUDIES FOR A LARGE HIGH TEMPERATURE GRAPHITE MODERATED REACTOR (1)

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ABSTRACT

Physics and optimisation studies of large (1250 MW; 10 MW/m³) Plutonium-fuelled reactors are carried out. Either uranium-238 or thorium is considered as fertile material. Complete core replacement is assumed.

1. — INTRODUCTION

Until now the Dragon Project has assessed fuel cycles for a large power reactor on the basis of U^{235} with an enrichement of 93 % and over a wide range of conditions.

Due to the progressive production of plutonium it seems to be advisable to study the substitution of enriched uranium by plutonium. In this report complete core replacement with no recycling has been assumed. Two systems, a thorium-plutonium and a U^{238}-plutonium reactor, are studied and optimised. The influence of self-shielding has been investigated.

2. — ASSUMED DATA

2.1. — BURN-UP DATA

All cases assume the following data :

TABLE 2.1.1. — Specifications of the Core

Thermal power output	1,250 MW
Average core power density	10 MW/m³
Moderator temperature	1,300 °K
Fuel temperature	1,500 °K
Buckling : taken from similar U^{235} cases	4.95×10^{-5} cm^{-2} for $S_{feed} = 2{,}000$ 5.64×10^{-5} cm^{-2} for $S_{feed} = 5{,}000$ (corresponds to approximately 2-3% leakage at start-up)
Graphite concentration	6.7×10^{22} atoms/cm³ (= 0.111 moles/cm³)
Nuclear data	Dragon Library II [2]
Fission product data	Ref. Dragon Yield II
Basic timestep (time between two spectrum calculations)	73 days
Loading procedure	Complete core replacement with no reprocessing

(1) Dragon Project Report 335, April 1965.

TABLE 2.1.2. — Feed Compositions

System	Feed Material	
	Fissile Material	Fertile Material
Th/Pu	"Civil plutonium"	Thorium-232
U^{238}/Pu	"Civil plutonium" + small amount of U^{235} due to natural uranium	U^{238} from natural uranium

TABLE 2.2.1. — Cost Data

Cost Items	Standard Prices Assumed
Purchase of fissile material	£5,000 per kg of fissile plutonium (Pu^{239} + Pu^{241})
Purchase of fertile material	£15 per kg thorium £14 per kg U^{238}
Fabrication of fuel particles	£150 per kg of heavy metal fabricated
Graphite cost	£1 per kg
Interest for above items from date of payment to date of assumed electricity return	6 % per annum interest rate (compound interest is calculated)
The following data are assumed :	
Date of payment	180 days prior to fuel insertion into the core
Date of electricity return	Assumed at half-time of a core-life (= cycle time)
Load factor	80 %
Net efficiency for electricity	40 %

Two systems have been studied having a range and a number of cases sufficient for optimisation purposes. They assume a homogeneous distribution of the feed materials in the core according to the table 2.1.2.

"Civil plutonium" results from an irradiation of 3,000 MWd/ton and has the following isotopic composition :

$$80\ \%\ \text{of}\ Pu^{239},\ 4\ \%\ \text{of}\ Pu^{241}\ \text{and}\ 16\ \%\ \text{of}\ Pu^{240}.$$

Natural uranium consists of 99.3 % U^{238} and 0.7 % U^{235}.

In addition some cases have been computed for a set of disadvantage factors to study the influence of fuel self-shielding on burn-up performance.

2.2. — Cost Analysis.

The cost analysis, carried out with the programme ELECTRA [9], takes the following items listed in table 2.2.1. into account. For optimisation purposes some of the data are also varied (especially the plutonium price). No credit is given for any fissile or fertile material present in the core at the end of the cycle.

3. — Optimisation of Plutonium-systems

3.1. — Presentation of Results.

One finds in Tables 3.1.1. to 3.1.4 the cases which have been studied for optimisation. The following results are listed : cycle time, conversion, fifa, cost (assuming standard prices) and the fissile and fertile amounts in terms of S_{feed} and N_{feed}. The amount of fissile material at the end is also given.

Cycle time, fifa, cost and conversion have been fitted each to an analytical function in S_{feed} and N_{feed} (rational function with 10 parameters).

The results are found in Figs. 3.1.1 to 3.1.4 for the Th/Pu system and in Figs. 3.1.5 to 3.1.8 for the U^{238}/Pu system.

A core composition which is situated below the critical line does not reach the normal end of life given by the depletion of all fissile materials but becomes under critical already in an early stage, due to the too high absorption in Pu^{240}

and other fertile materials, before the reactivity builds up again due to destruction of Pu^{240} and production of Pu^{241}. There are ways to avoid this such as lumping the fuel or zoning the core. The critical line refers to reactors which are just critical ($k_{eff} = 1.0$) after Xe and Sm equilibrium poisoning.

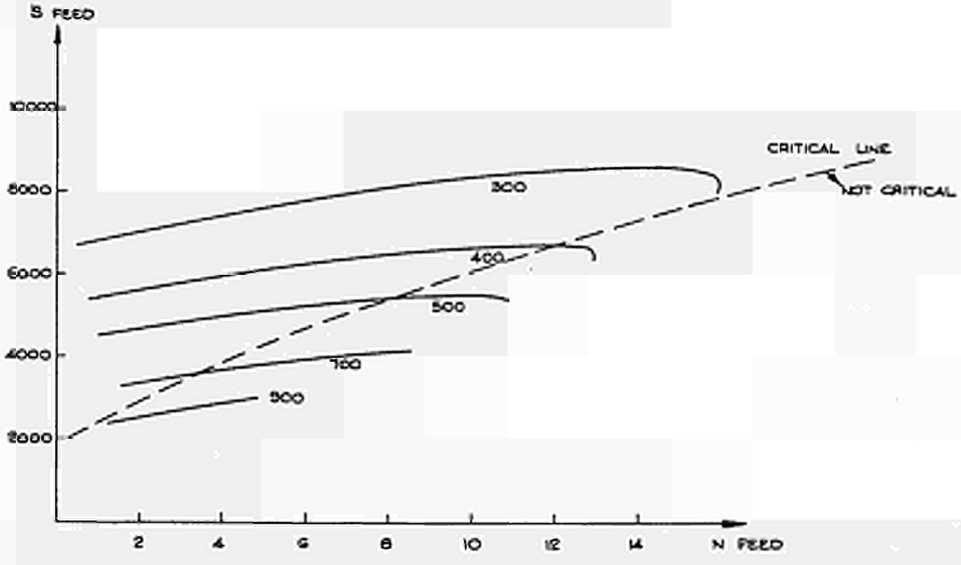


FIG. 3.1.1. — Cycle-Time (in Days) for a Th/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

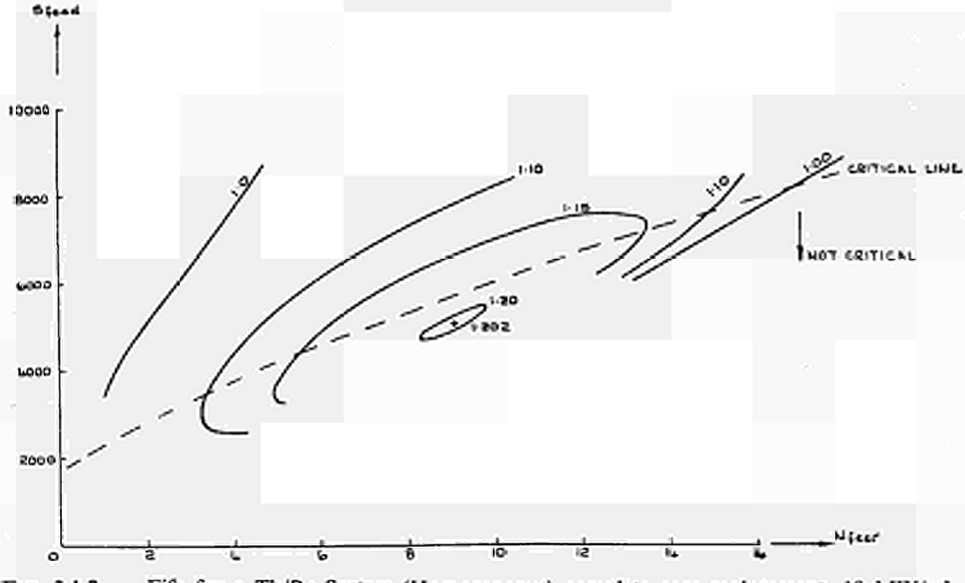


FIG. 3.1.2. — Fifa for a Th/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

TABLE 3.1.1. — Cycle Time, Fifa, Conversion and Cost for Th/Pu Systems

S_{feed} (Pu)		Th/Pu System (Homogeneous) Complete Core Replacement, 10 MW/m³, No Reprocessing						
	N_{feed} (Pu)	0	0.3					
2,000	Cycle time	1,033	1,062					
	Fifa	0.901	0.926					
	Conversion	0.005	0.047					
	Cost (d/kWh)	0.1941	0.1909					
	N_{feed} (Pu)	0	3	4				
4,000	Cycle time	519	613	639				
	Fifa	0.905	1.069	1.114				
	Conversion	0.007	0.245	0.310				
	Cost (d/kWh)	0.1871	0.1739	0.1718				
	N_{feed} (Pu)		2	4	7			
5,000	Cycle time		459	494	537			
	Fifa		1.00	1.081	1.170			
	Conversion		0.149	0.280	0.426			
	Cost (d/kWh)		0.1795	0.1760	0.1762			
	N_{feed} (Pu)	0			7	10		
6.000	Cycle time	351			431	454		
	Fifa	0.921			1.131	1.191		
	Conversion	0.001			0.399	0.511		
	Cost (d/kWh)	0.1840			0.1820	0.1859		
	N_{feed} (Pu)					10	14	
8,000	Cycle time					318	325	
	Fifa					1.109	1.133	
	Conversion					0.454	0.570	
	Cost (d/kWh)					0.1994	0.2127	
	N_{feed} (Pu)	0		5			14	18
10,000	Cycle time	216		230			245	239
	Fifa	0.941		1.002			1.068	1.042
	Conversion	0.001		0.240			0.524	0.621
	Cost (d/kWh)	0.1840		0.1974			0.2268	0.2511

N. B. — Costs are for standard prices, e.g., £5,000/kg fissile Pu
£150/kg heavy metal fabricated
£15/kg thorium
£1/kg graphite
6% interest per annum
The cases with N = 0 have been abandoned for optimisation since the accuracy is not very good at the end of life.

TABLE 3.1.2. — S_{feed} ,N_{feed} S_{final} for Th/Pu Systems

S_{feed} (Pu)		Th/Pu System (Homogeneous) Complete Core Replacement 10 MW/m³, No Reprocessing						
	N_{feed} (Pu)	0	0.3					
2,000	S_{feed}	2,000	2,000					
	N_{feed}	0.03	0.3					
	S_{final}	39,000	37,000					
	N_{feed} (Pu)	0	3	4				
4,000	S_{feed}	4,000	4,000	4,000				
	N_{feed}	0.06	3	4				
	S_{final}	57,900	30,000	24,000				
	N_{feed} (Pu)		2	4	7			
5,000	S_{feed}		5,000	5,000	5,000			
	N_{feed}		2	4	7			
	S_{final}		54,000	32,500	20,200			
	N_{feed} (Pu)	0			7	10		
6,000	S_{feed}	6,018			6,018	6,018		
	N_{feed}	0.01			7.02	10.03		
	S_{final}	138.000			26,000	18,800		
	N_{feed} (Pu)					10	14	
8,000	S_{feed}					8,000	8,000	
	N_{feed}					10	14	
	S_{final}					27,700	20,000	
	N_{feed} (Pu)	0		5			14	18
10,000	S_{feed}	10,000		10,000			10,000	10,000
	N_{feed}	0.01		5			14	18
	S_{final}	200,000		61,000			28,000	22,000

TABLE 3.1.3. — Cycle Time, Fifa, Conversion and Cost for U^{238}/Pu Systems

S_{feed} (Pu)		U^{238}/Pu System (Homogeneous) Complete Core Replacement, 10 MW/m³, No Reprocessing						
	N_{feed} (Pu)	0						
2,000	Cycle time	1,033						
	Fifa	0.901						
	Conversion	0.005						
	Cost (d/kWh)	0.1941						
	N_{feed} (Pu)	0	1					
4,000	Cycle time	519	649					
	Fifa	0.905	1.124					
	Conversion	0.007	0.205					
	Cost (d/kWh)	0.1871	0.1599					
	N_{feed} (Pu)		1	2	3			
5,000	Cycle time		521	578	619			
	Fifa		1.128	1.242	1.321			
	Conversion		0.194	0.311	0.401			
	Cost (d/kWh)		0.1548	0.1445	0.1393			
	N_{feed} (Pu)	0	1	2	3	4		
6,000	Cycle time	351	413	462	515	533		
	Fifa	0.921	1.076	1.195	1.323	1.360		
	Conversion	0.001	0.165	0.279	0.377	0.444		
	Cost (d/kWh)	0.1840	0.1620	0.1498	0.1389	0.1382		
	N_{feed} (Pu)			2	3	5	7	
8,000	Cycle time			336	371	407	416	
	Fifa			1.155	1.267	1.371	1.392	
	Conversion			0.246	0.335	0.451	0.501	
	Cost (d/kWh)			0.1556	0.1453	0.1401	0.1407	
	N_{feed} (Pu)	0			3	5	7	9
10,000	Cycle time	216			290	314	334	311
	Fifa	0.941			1.238	1.322	1.388	1.276
	Conversion	0.001			0.314	0.413	0.499	0.566
	Cost (d/kWh)	0.1840			0.1500	0.1462	0.1447	0.1625
	N_{feed} (Pu)						7	11
12,000	Cycle time						279	244
	Fifa						1.395	1.188
	Conversion						0.475	1.188
	Cost (d/kWh)						0.1453	0.1813
	N_{feed} (Pu)						9	15
15,000	Cycle time						226	151
	Fifa						1.391	0.895
	Conversion						0.508	0.620
	Cost (d/kWh)						0.1534	0.2569

N. B. The same data are assumed as in Table 3.1.1.

TABLE 3.1.4. — S_{feed}, N_{feed}, S_{final} for U^{238}/Pu Systems

S_{feed} (Pu)		U^{238}/Pu System (Homogeneous) Complete Core Replacement, 10 MW/m³, No Reprocessing						
	N_{feed} (Pu)	0						
2,000	S_{feed}	2,000						
	N_{feed}	0.03						
	S_{final}	39,000						
	N_{feed} (Pu)	0	1					
4,000	S_{feed}	4,000	3,972					
	N_{feed}	0.06	0.99					
	S_{final}	57,900	66,700					
	N_{feed} (Pu)		1	2	3			
5,000	S_{feed}		4,965	4,931	4,897			
	N_{feed}		0.99	1.97	2.94			
	S_{final}		92,700	55,600	33,000			
	N_{feed} (Pu)	0	1	2	3	4		
6,000	S_{feed}	6,018	5,976	5,935	5,894	5,854		
	N_{feed}	0.01	1.00	1.98	2.95	3.9		
	S_{final}	138,000	97,000	66,700	50,000	32,000		
	N_{feed} (Pu)			2	3	5	6	
8,000	S_{feed}			7,890	7,836	7,730	7,678	
	N_{feed}			1.97	2.94	4.82	5.76	
	S_{final}			96,300	75,260	41,900	31,900	
	N_{feed} (Pu)	0			3	5	7	6
10,000	S_{feed}	10,000			9,795	9,663	9,533	9,410
	N_{feed}	0.01			2.94	4.83	6.67	8.47
	S_{final}	200,000			102,000	60,000	40,000	24,200
	N_{feed} (Pu)						7	11
12,000	S_{feed}						11,473	11,172
	N_{feed}						6.68	10.24
	S_{final}						58,303	25,200
	N_{feed} (Pu)						9	15
15,000	S_{feed}						14,117	13,690
	N_{feed}						8.62	13.54
	S_{final}						58,300	22,400

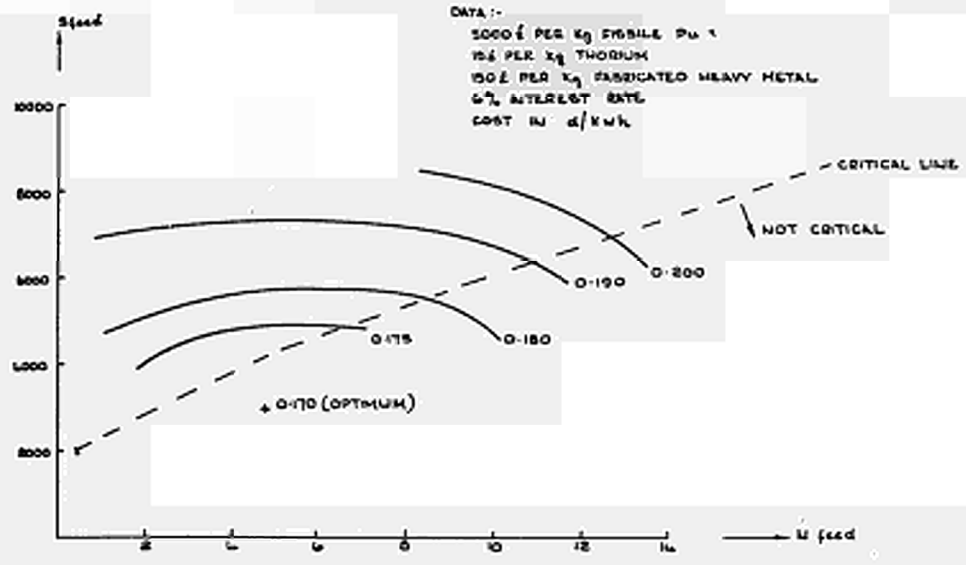


FIG. 3.1.3. — Cost for a Th/Pu System (Homogeneous complete) core replacement, 10 MW/m³, no reprocessing.

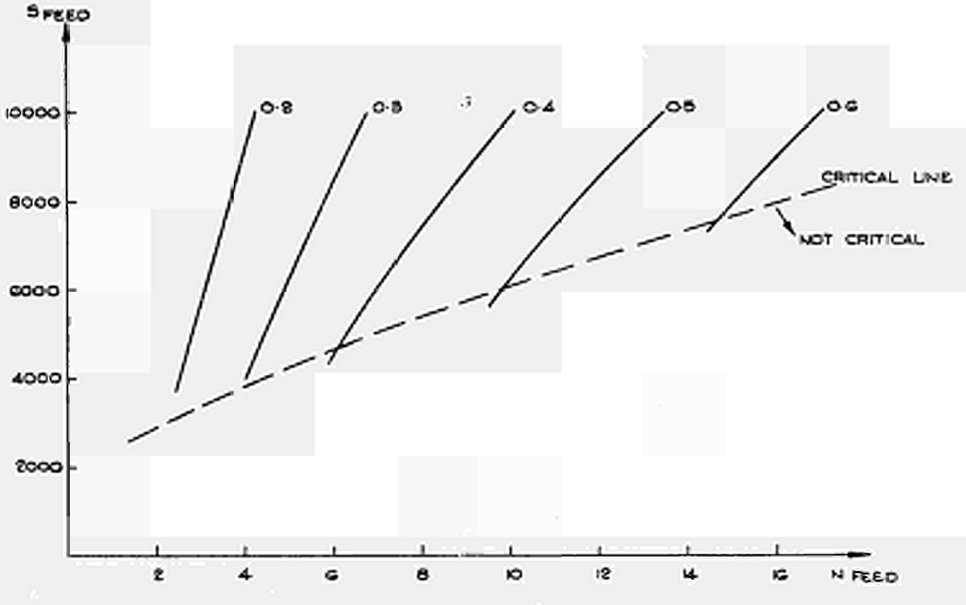


FIG. 3.1.4. — Conversion for a Th/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

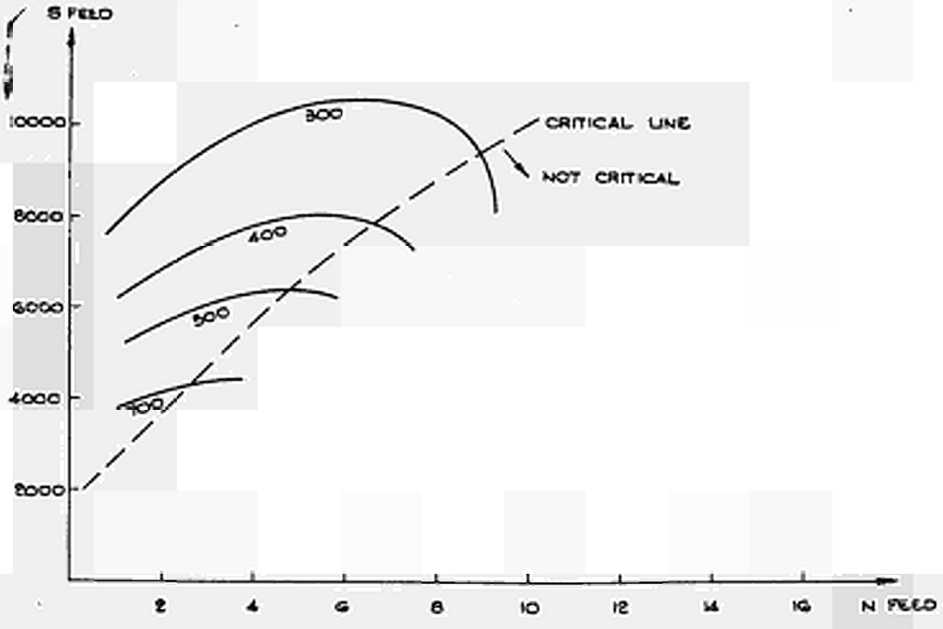


FIG. 3.1.5. — Cycle Time (Days) for a U^{238}/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

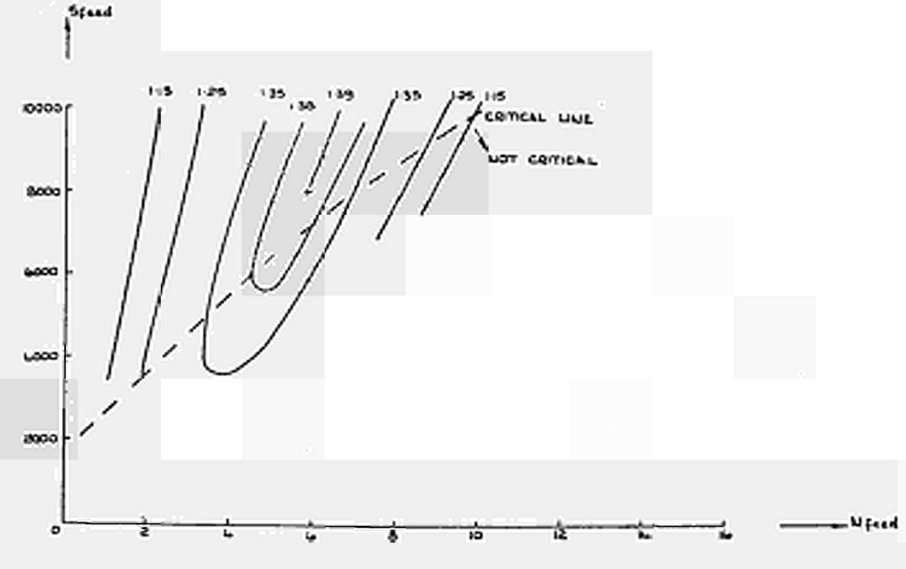


FIG. 3.1.6. — Fifa for a U^{238}/Pu System (Homogeneous) complete core replacement, no reprocessing.

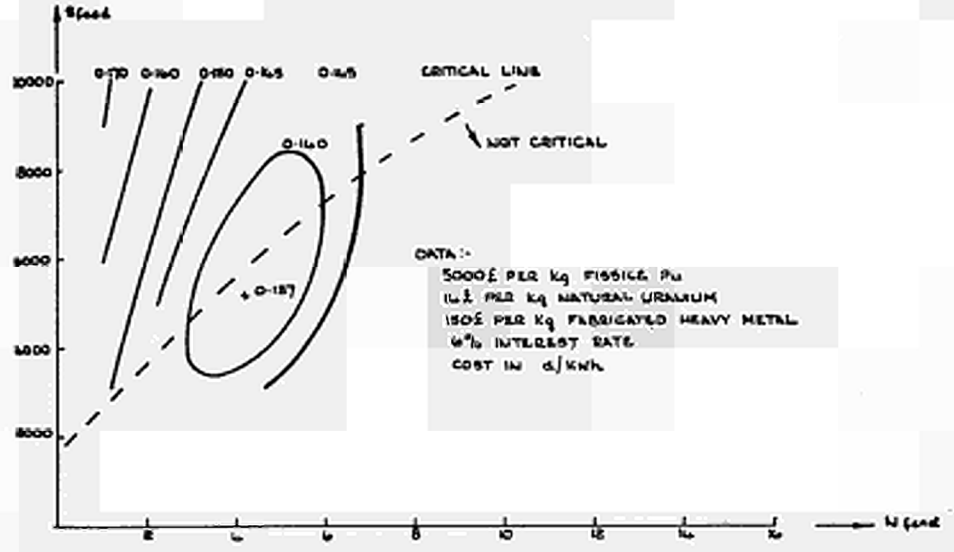


FIG. 3.1.7. — Cost for a U^{235}/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

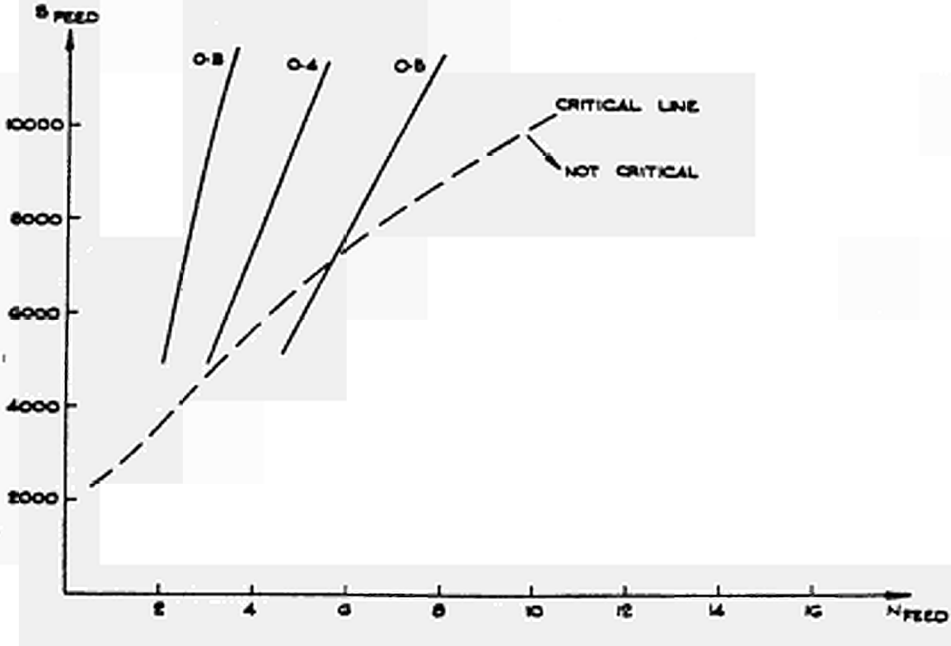


FIG. 3.1.8. — Conversion for a U^{235}/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

3.2. — Main Characteristics.

3.2.1. — *Cycle Time.*

For constant plutonium-investment the increase of fertile material content increases the cycle time owing to a better conversion which slows down the reactivity drop at the end of life. An adverse effect is the decrease of the multiplication factor at start-up, which tends to shorten the core life. The former effect is more significant in U^{238} systems than in thorium systems owing to the production of Pu^{239} instead of U^{233}. The former has a very much higher fission cross section and, therefore, has the same probability to be fissioned as the originally inserted fissile material. In the case of U^{233} being produced from thorium the probability for a U^{233} atom to be fissioned is approximately three times less than for a Pu^{239} atom. In addition Pu^{239} is less sensitive to fission product poisoning.

The life of the core is roughly proportional to the initial amount of fissile material.

3.2.2. — *Fifa Value.*

A similar argument as above applies for constant plutonium-investment, but for a constant N-value fifa has a relative maximum when varying S. Two opposite effects explain this behaviour. A hard spectrum has better conversion properties but the decrease in capture to fission rate in Pu^{239} with increasing S-value more than compensates this effect. Thus the fifa value increases. For high S-values fifa tends to decrease again due to the increasing relative losses in parasitic absorptions and leakage. Hence a maximum in the (S_f, N_f) field exists. It is interesting to note that compared to a U^{235} system of a similar kind the mountain is much flatter and especially the influence of the spectrum is much less important (ridge for U^{238} systems). A Pu/U^{238} system gives higher fifa values than a Pu/Th system.

3.2.3. — *Cost.*

Similar arguments and properties can be applied as for fifa, but the position of the optimum depends on the fabrication cost as well (as described in 3.3). Thorium cycles are more expensive than U^{238} cycles. Note again the smaller influence of the S-value in the latter systems.

3.2.4. — *Conversion.*

For S = constant the increase of fertile material improves the conversion. For N = constant the conversion benefits from a harder spectrum. The resonance integral of U^{238} being approximately three times larger than the one for Th^{232}, the conversion of a U^{238} system is higher for the same N-value.

N. B. — The values are taken from the programme HELIOS, which does not include Pu^{240} when calculating the average cycle conversion.

3.3. — Optimum Reactor for Different Price Parameters.

For a set of price parameters a cost analysis of the two mentioned systems has been performed.

Tables 3.3.1 and 3.3.2 show the assumed price data, the optimum core position and the optimum fuel cost with these price data. Figs. 3.3.1 to 3.3.4 show the results on graphs.

For negligible fabrication cost the position of fifa maximum determines approximately the best reactor, but with an increase of the fabrication charge a shift to lower fertile material concentrations is more economic.

A cheaper plutonium price favours more undermoderated reactors with less conversion.

High fuel element cost (graphite component) requires reactors with higher fissile investment.

The plutonium price is by far the largest contribution to the electricity cost. Therefore, the latter are very sensitive to the assumption made for plutonium charges and to a minor degree only to variations in fabrication cost and costs for fertile and moderator materials.

4. — Physics of some Selected Cases

4.1. — Presentation of Results.

Three cases have been selected with the aim of understanding the physics of a plutonium system :

(i) a homogeneous thorium/plutonium core,
(ii) a homogeneous U^{238}/plutonium core and
(iii) a self-shielded U^{238}/plutonium core.

The burn-up performances of these reactors are not far from their respective optimum.

4.2. — Main Characteristics.

(i) *Neutron Balance* (Fig. 4.2.1).

The f-value reflecting the neutron loss in materials other than heavy metals drops sharply at the beginning owing to Xe and Sm and continues to decrease as a result of the build-up of fission products.

The η_{HM}-value increases during life for the considered homogeneous systems, because the fission rate of Pu^{239} decreases while the fission rate of Pu^{241} increases; Pu^{241} has a better η-value than Pu^{239}. In addition the Pu^{240} absorption rate decreases. In the shielded case the Pu^{240} effective cross section is lower and hence Pu^{240} builds up at least at the beginning. The slower Pu^{241} conversion and the higher absorption in Pu^{240} causes the different shape of η_{HM}.

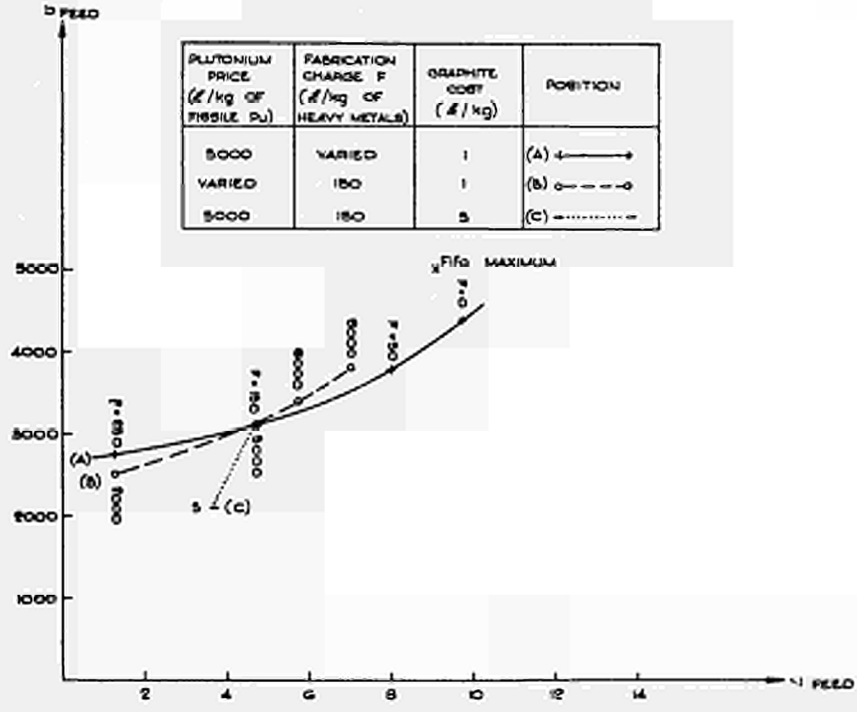


FIG. 3.3.1. — Composition of optimum reactor for a Th/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

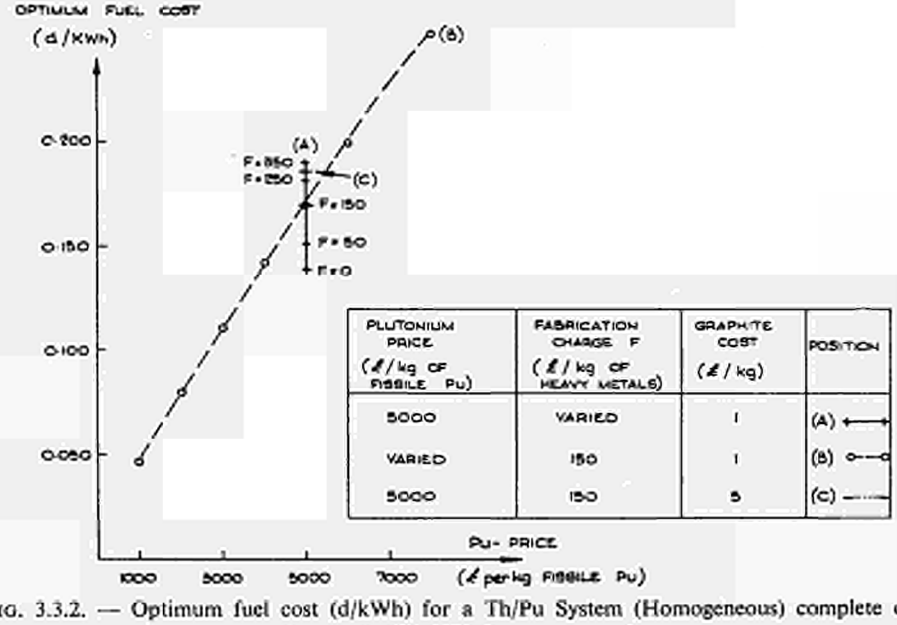


FIG. 3.3.2. — Optimum fuel cost (d/kWh) for a Th/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

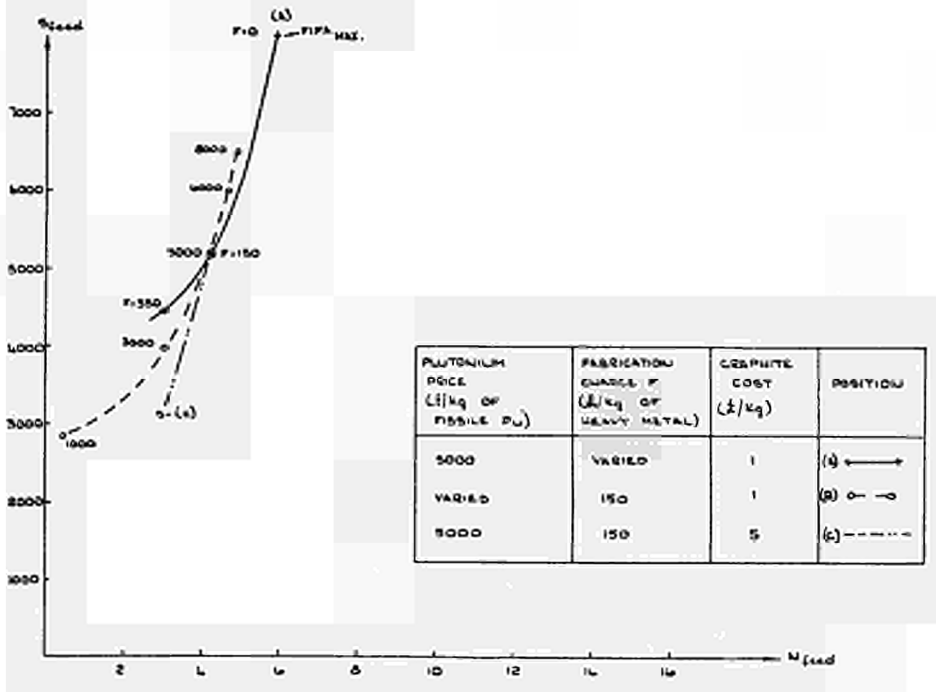


FIG. 3.3.3. — Composition of optimum reactor for U^{238}/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

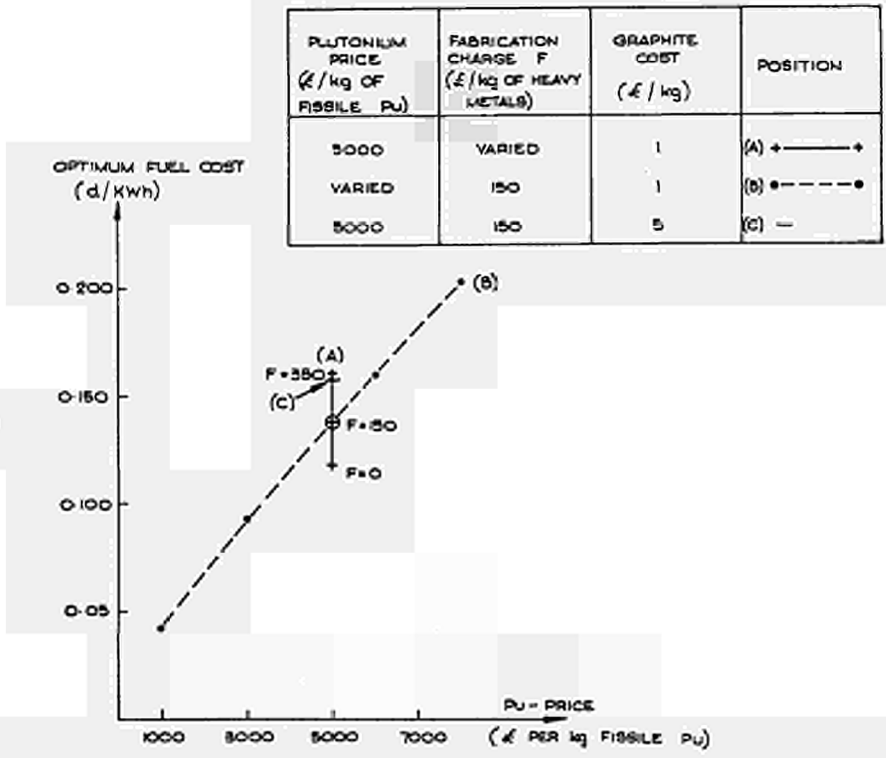


FIG. 3.3.4. — Optimum fuel cost (d/kWh) for a U^{238}/Pu System (Homogeneous) complete core replacement, 10 MW/m³, no reprocessing.

TABLE 3.3.1. — Optimised Reactor : Th/Pu System

Complete Core Replacement, 10 MW/m³, No Reprocessing						
In Respect of	Fabrication Charge (£/kg HM)	Plutonium Price Fissile Pu) (£/kg	Fuel Element (£/kg Graphite)	Position		Fuel Cost Optimum (d/kWh)
				S_{feed}	N_{feed}	
Cost Minimum	0	5,000	1.0	4,400	9.7	0.139
	50	5,000	1.0	3,800	8.0	0.151
	150	5,000	1.0	3,100	4.7	0.170
	250	5,000	1.0	2,750	1.25	0.181
	350	5,000	1.0	2,780	1.20	0.190
	150	1,000	1.0	2,000	1.10	0.047
	150	2,000	1.0	2,050	1.15	0.080
	150	3,000	1.0	2,500	1.25	0.111
	150	4,000	1.0	2,900	2.90	0.142
	150	6,000	1.0	3,400	5.7	0.198
	150	8,000	1.0	3,650	6.9	0.252
	150	5,000	5	2,100	3.7	0.186
Fifa Maximum = 1.20				5,000	9.0	
An optimised reactor is understood to be the best reactor system under the given conditions in the (S_f, N_f) field.						

TABLE 3.3.2. — Optimised Reactor : U^{238}/Pu System

Complete Core Replacement, 10 MW/m³, No Reprocessing						
In Respect of	Fabrication Charge (£/kg HM)	Plutonium Price (£/kg Fissile Pu)	Graphite Price (£/kg Graphite)	Position		Fuel Cost Optimum (d/kWh)
				S_{feed}	N_{feed}	
Cost Minimum	0	5,000	1	8,000	5.9	0.118
	150	5,000	1	5,200	4.2	0.138
	350	5,000	1	4,400	3.0	0.160
	150	5,000	5	3,200	3.0	0.158
	150	1,000	1.0	2,850	0.5	0.042
	150	3,000	1.0	4,000	3.0	0.092
	150	6,000	1.0	6,000	4.6	0.160
	150	8,000	1.0	6,500	5.0	0.202
	Variation of U^{238} Price			Negligible Effect on Position and Cost		
Fifa Maximum = 1.39				8,000	6.0	
Optimum reactor is understood to be the best reactor system under the given conditions in the (S_f, N_f) field.						

The resonance escape probability does not change much during the core life reflecting the negligible burn-up of U^{238} respective Th^{232}.

The leakage in a big power reactor is not very important (approximately 2-3 % at start-up).

(ii) K_{eff} (Fig. 4.2.1).

K_{eff} results from the competition between the continuous worsening of f and the possible improvement of η_{HM}. The reactivity curve drops after start-up due to rapid poisoning (sharp decrease of f). When the saturating fission products and heavy isotopes (e.g., Pa^{233}) have reached equilibrium, the smoother decrease of f may be overcome by the increase of η_{HM} which effects a possible rise of k_{eff}. The steep decrease of reactivity at the end of life is due to a high relative depletion of fissile isotopes. The reactivity curve is flatter for reactors with more fertile material. The self-shielding similarly effects the reactivity curve.

(iii) *Neutron Flux.*

Fig. 4.2.2 represents the spectrum using a four-group structure. The plotted curve (e.g., 3 eV) represents the ratio between the integrated neutron flux from 0 up to the assigned energy and the total flux. The dotted line is the total flux in absolute units.

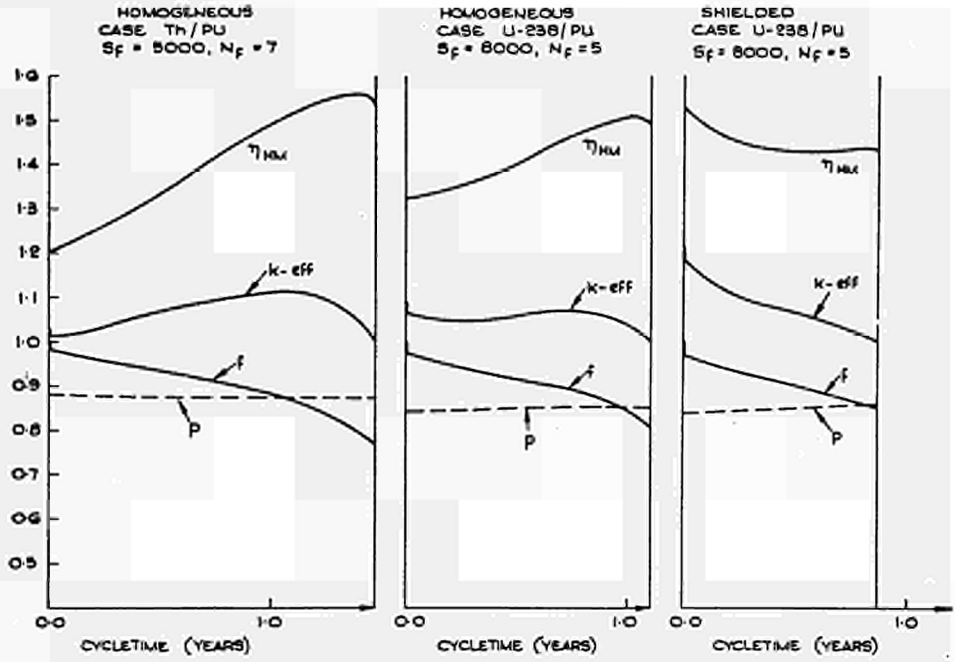


FIG. 4.2.1. — Neutron Balance for selected Cases.

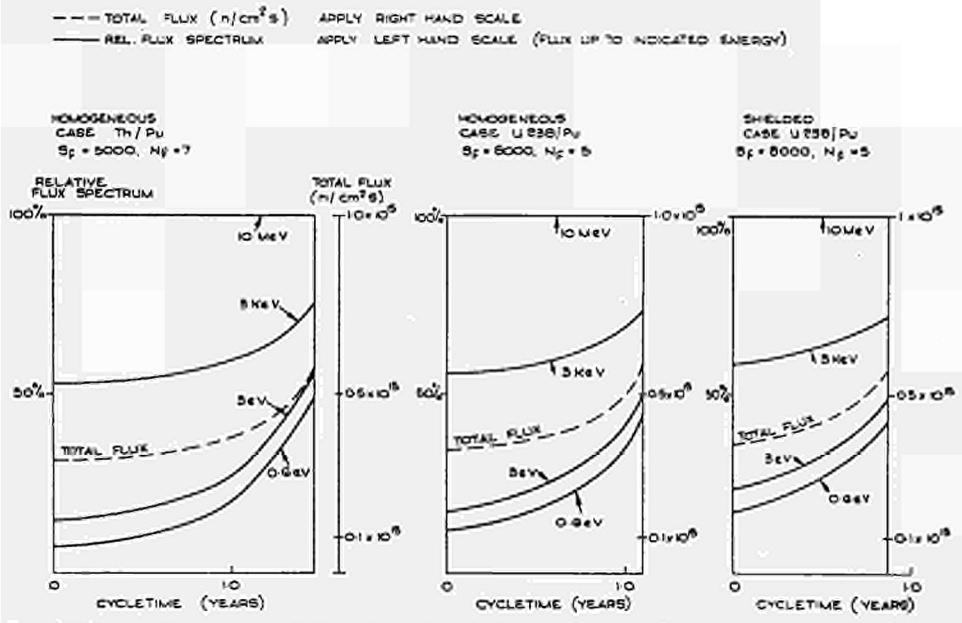


FIG. 4.2.2. — Spectrum and flux for selected Cases complete Core Replacement, 10 MW/m³, no Reprocessing.

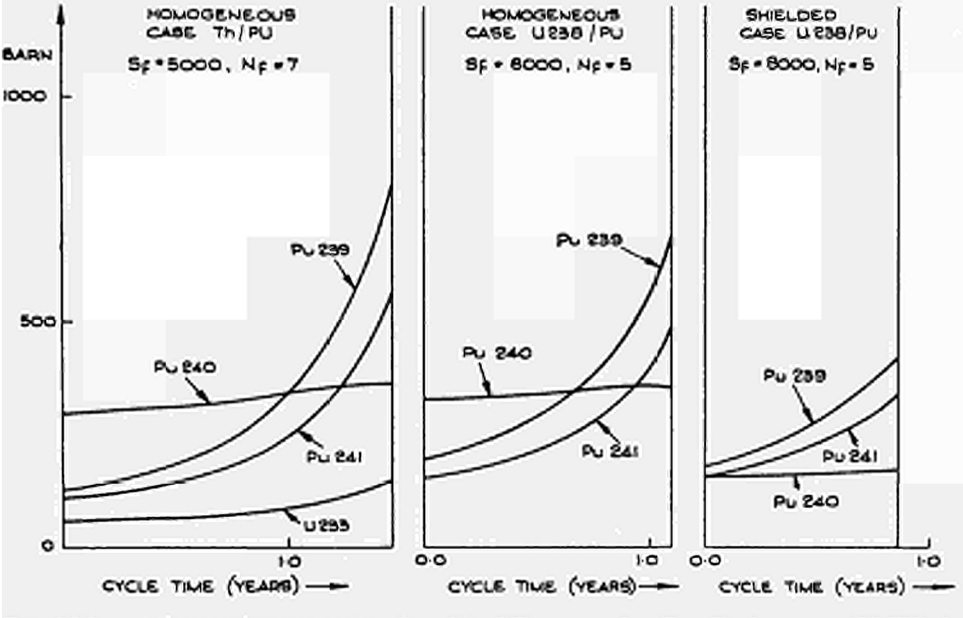


FIG. 4.2.3. — Microscopic cross section for selected Cases complete Core Replacement, 10 MW/m³, no Reprocessing.

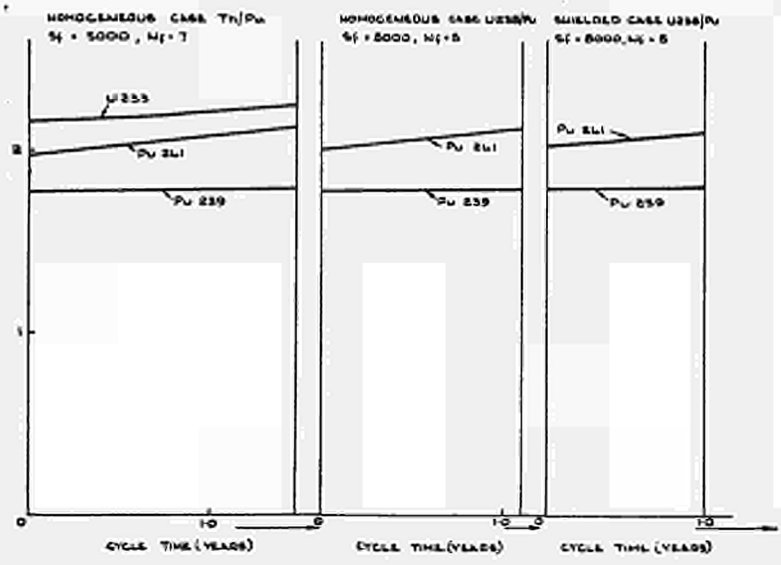


FIG. 4.2.4. — Eta for selected Cases, complete Core Replacement, 10 MW/m³, no Reprocessing.

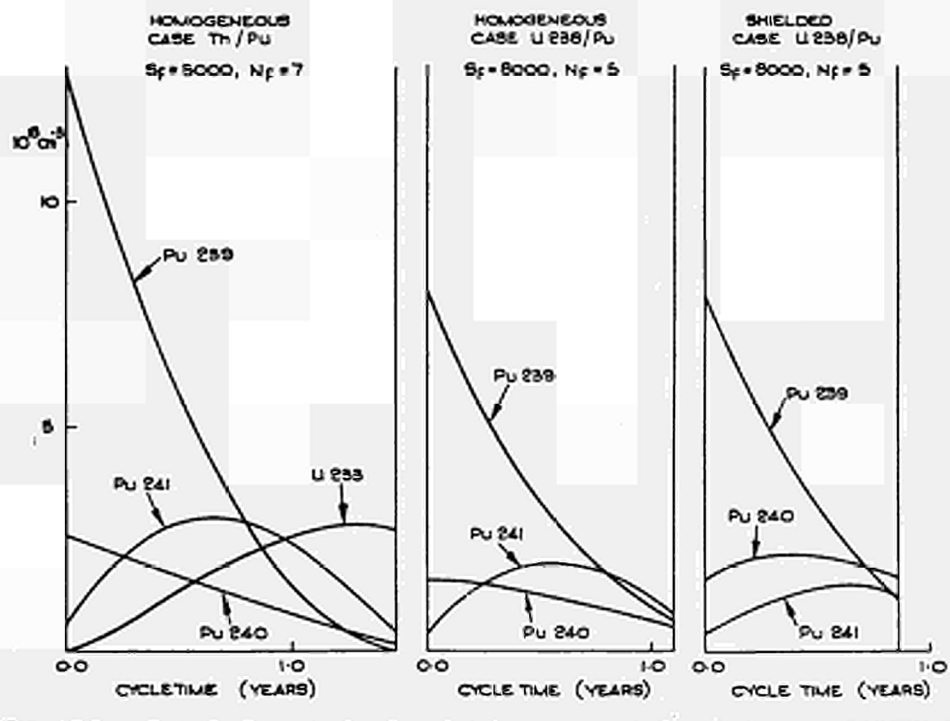


FIG. 4.2.5. — Isotopic Concentration for selected cases complete Core Replacement, 10 MW/m³, no Reprocessing.

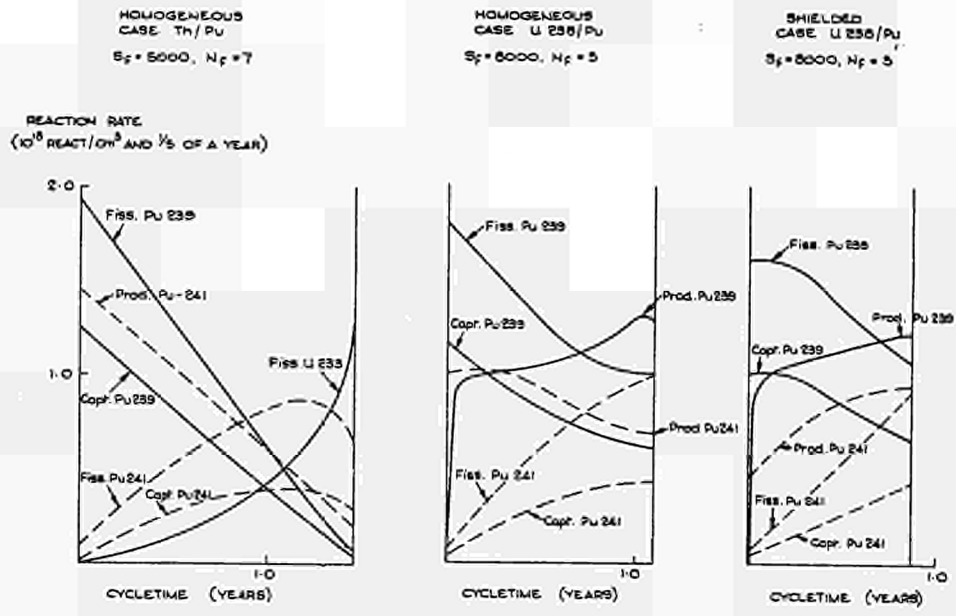


FIG. 4.2.6. — Reaction Rate for Pu-Chain of some selected Cases complete Core Replacement, 10 MW/m³, no Reprocessing.

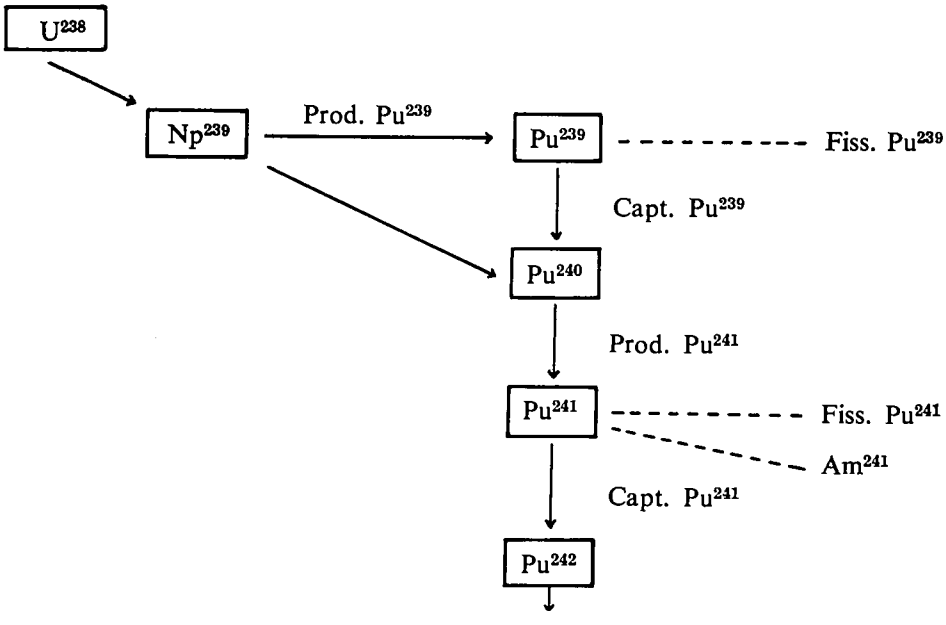
The fast flux remains constant during the life since the reactor runs on constant power. The thermal flux increases owing to the depletion of fissile material and the constant power condition.

(iv) *Build-up and Depletion of some Isotopes* (Fig. 4.2.5).

Pu^{239} is depleted during life by fission and neutron capture. The latter one produces Pu^{240} which is converted into Pu^{241}. Pending on the balance between production and destruction, the Pu^{240} will either build-up or decrease; in a not too soft spectrum with no-self-shielding and the currently considered Pu^{240} grade the destruction prevails. Pu^{241} builds up at the beginning and decreases after some time. This is because in the later stage less Pu^{239} is available and hence the Pu^{241} has to provide the fissions which are required to maintain the constant power level. In addition the Pu^{241} production decreases. The insertion of fertile material will slow down the depletion of the fissile isotopes.

From the concentrations of the various isotopes, the one-group flux and the one-group microscopic cross section (Fig. 4.2.3) it is easy to infer the most impor-

tant reaction rates in the systems considered (Fig. 4.2.6). The following simplified model has been used for the plutonium chain :



5. — Influence of the Self-Shielding on the Physics and Burn-up Performance of a Plutonium-System

Although no systematic investigation and optimisation has been done, some general advantages and rules can be given. Three cases using U^{238} as fertile material are presented as listed in Table 5.1.

Table 5.1. — Cases Considered in the Study of the Influence of Self-Shielding

Complete Core Replacement 10 MW/m³		Parameters for Self-Shielding (particle diameter) (cm)	Disadvantage Factors		
			HELIOS Group		
S_{feed} (Pu)	N_{feed} (Pu)		20	21	22
5,000	3	0.00 (homog.)	1.44	0.84	1.40
5,000	3	0.05	1.21	0.40	1.14
5,000	4	0.05	1.25	0.45	1.18
		Energy boundaries 0.825 — 1.0 — 1.1 — 1.3 (eV)			

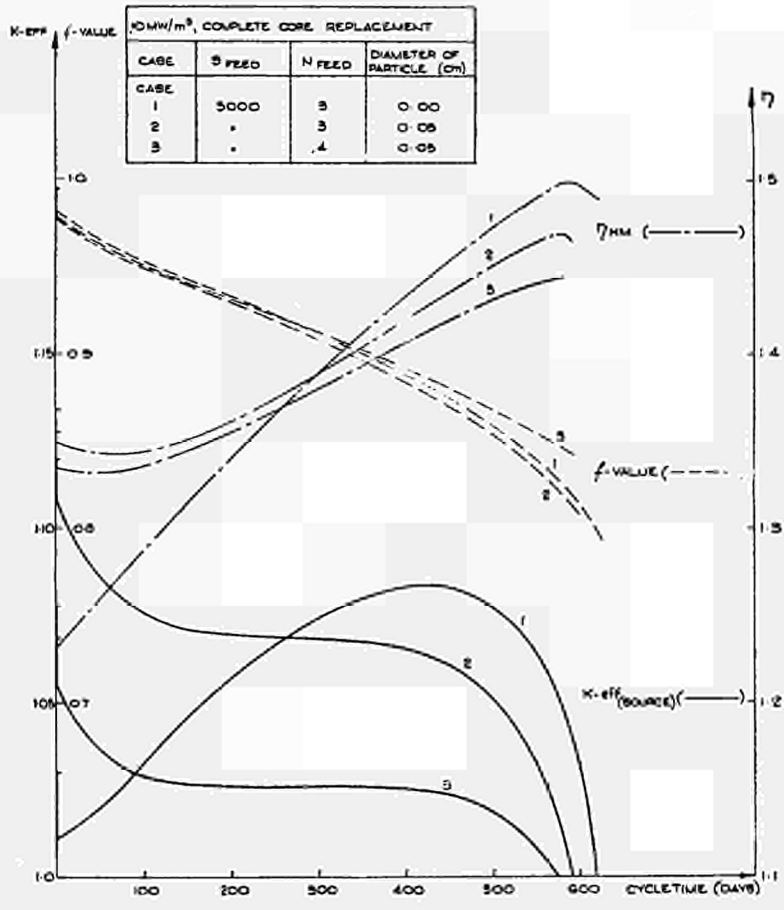


FIG. 6.1. — Influence of self shielding on the physics of a Pu-U^{238} system complete core replacement, 10 MW/m³, no reprocessing.

Fig. 6.1 shows the neutron balance during life.

The self-shielding decreases the effective Pu^{240} cross section more than those of the other materials owing to the high resonance integral of Pu^{240} at 1.057 eV. This is reflected in the higher η_{HM} value at start-up (less neutrons are lost in Pu^{240}).

During burn-up the destruction of Pu^{240} is held up which increases the Pu^{240} pile up since the production rate of Pu^{240} is in both cases nearly the same. So progressively more neutrons are lost in Pu^{240} in spite of its smaller cross section. Also the improvement in the fissile η-value which results from the switching over from Pu^{239} fissions to Pu^{241} fissions is delayed by self-shielding. These two effects make the shape of η_{HM} flatter and its value smaller at the end of life.

The f-value is in both cases nearly the same. The non-heavy metal absorption is equal but the heavy metal absorption is less in the shielded case. Hence the re-

lative non-heavy metal absorption is more dominant; this causes the slight difference.

The k_{eff} curve follows $\eta_{HM} \cdot f$.

TABLE 5.2. — Influence of Self-Shiedling on Burn-up

S_{feed} (Pu)	N_{feed} (Pu)	Diameter (cm)	Cycle Time (days)	Comparison at Time 585 days		
				Fissile Pu (10^{18} cm^{-3})	Fraction of Pu^{239}	U^{238} (10^{19} cm^{-3})
5,000	3	0.00	618	2.65	44 %	3.29
5,000	3	0.05	591	2.44	44 %	3.28

In the shielded case more fissions come from Pu^{239} in the first stage of core life. So the rate of depletion of fissile material necessary to give the same power is higher owing to the larger α of Pu^{239}. The U^{238} conversion is the same in both cases. In addition to the higher depletion the neutron economy is less favourable so that the self-shielded system becomes already subcritical at a higher fissile concentration. These two effects shorten the core life.

One can utilise the almost flat behaviour of k_{eff} in the self-shielded case to lower the control requirements. First there is a somewhat lower demand in total control anyway (see Fig. 6.1 curves 1 and 3). This difference will be even more marked for lower initial S-values. Second the self-shielded case has its highest k_{eff} initially, where it is controllable by burnable poison, whereas the homogeneous case must be controlled by movable control units.

It is certainly possible to select the self-shielding parameter so as to get the best control characteristics and a reasonable burn-up performance. In reality the self-shielding will vary during life which will somehow change the previous burn-up characteristics.

6. — CONCLUSIONS AND SUMMARY

Table 6.1 summarises the most important results, e.g., fifa and cost. The two plutonium-systems studied are given and compared with optimised uranium systems. If no reprocessing facilities are available for U^{233}-U^{235}-thorium fuel, the fuel costs for a batch loaded Pu/U^{238} fuel cycle are equal to those of a U^{235}/thorium cycle with yearly replacement of one quarter of the core, assuming the charges for fissile and fertile materials and fabrication respectively are the same in both cases.

A batch loaded U^{235}/thorium cycle (complete core replacement) is less interesting than the Pu/U^{238} cycle. However, in the latter case one cannot expect to gain

much by zoning or reprocessing, since nearly all the fissile plutonium is already consumed at the end of life of a simple batch loaded cycle.

A thorium/plutonium system is less advantageous than a U^{238}/Pu system. But one should be careful not to generalise and apply these results to other systems such as seed and blanket or segregated fuel .

Although no systematic investigation has been done on the effect of self-shielding it seems that fuel lumping is interesting, mainly because of improvement in the control characteristics of the system.

TABLE 6.1. — Main Optimum Results

Optimum Reactor in respect of	Pu Systems Complete Core Replacement No Reprocessing		Th/Highly Enriched U^{235} Systems	
	Th/Pu	U^{238}/Pu	Complete Core Replacement No Reprocessing	4-Zone Core No Reprocessing
Fifa maximum	1.20	1.39	1.12	1.60
Cost minimum (d/kWh) (standard prices) . . .	0.170	0.137	0.185	0.135

N. B. — Reactor with Fifa maximum does not coincide with optimum cost reactor.

Standard prices are : £5,000 for U^{235} (93 % enriched)
£5,000 for fissile plutonium (civil grade)
£150 fabrication charge per kg heavy metal
£150 separation charge per kg heavy metal
£15 per kg thorium
£14 per kg U^{238}
£1 per kg graphite
6 % interest rate

7. — A LIST OF SYMBOLS AND RELATIONS USED IN THIS PAPER

k_{eff} (source) effective multiplication factor according to the source iteration procedure (spectrum without control poison).

k_{eff} effective multiplication factor resulting from a spectrum in a core with control poison.

(1) $k_{eff} = \eta_{HM} \cdot f \cdot p \cdot L$

with

$$(1a)\ \eta_{HM} = \frac{\sum_{i=1}^{12} \nu\sigma_f^{(i)} \cdot N_i}{\sum_{i=1}^{12} \sigma_a^{(i)} \cdot N_i}$$

η_{HM} modified η-value for the following 12 isotopes : Th^{232}, Pa^{233}, U^{233}, U^{234}, U^{235}, U^{236}, U^{238}, Np^{239}, Pu^{239}, Pu^{240}, Pu^{241} and Pu^{242}.

$\nu\sigma_f(i)$ one-group neutron production cross section of the isotope i.

$\sigma_a(i)$ one-group neutron absorption cross section of the isotope i excluding resonance absorption in Th^{232}, U^{238}.

N_i concentration of isotope i.

$$(1b)\ f = \frac{\sum_{i=1}^{12} \sigma_a^{(i)} N_i}{\sum_{i=1}^{KMAT} \sigma_a^{(i)} N_i}$$

f fractional absorption in heavy metals to total absorption excluding Th^{232} and U^{238} resonances.

KMAT total number of isotopes treated in HELIOS (the sum does not include the control poison added to make the reactor critical).

$$(1c)\ p = \frac{\sum_{i=1}^{KMAT} \sigma_a^{(i)} N_i}{\sum_{i=1}^{KMAT} \sigma_a^{(i)} N_i + \sigma_{res}^{Th} N_{Th} + \sigma_{res}^{U^{238}} N_{Th}}$$

p resonance escape probability (only Th^{232} and U^{238}).

L non-leakage probability.

The one-group cross sections are condensed over a spectrum where control poison is in the core.

$(1d)\ \eta = \frac{\nu\sigma_f^{(i)}}{\sigma_a^{(i)}}$ conventional η-value of fissile isotopes.

α ratio of capture to fission cross section.

ν neutrons produced per fission.

$$S_{feed}^{(Pu)} = \frac{\text{moles of graphite loaded}}{\text{molcs of } (Pu^{239} + Pu^{241}) \text{ loaded}}$$

$$S_{feed} = \frac{\text{moles of graphite loaded}}{\text{moles of } (Pu^{239} + Pu^{241} + U^{233} + U^{235}) \text{ loaded}}$$

$$N_{feed}^{(Pu)} = \frac{\text{moles of } (Th + U^{238}) \text{ loaded}}{\text{moles of } (Pu^{239} + Pu^{241}) \text{ loaded}}$$

$$N_{feed} = \frac{\text{moles of } (Th + U^{238}) \text{ loaded}}{\text{moles of } (Pu^{239} + Pu^{241} + U^{233} + U^{235}) \text{ loaded}}$$

$$S_{final} = \frac{\text{moles of graphite at shutdown}}{\text{moles of } (Pu^{239} + Pu^{241} + U^{233} + U^{235}) \text{ at shutdown}}$$

cycle time in full power days.

fifa total number of fissions per fissile atom loaded at start-up.

conversion ratio of production of fissile material to the total destruction of fissile material during one cycle; it is calculated by

$$\text{conversion} = \frac{\Delta\text{Th} + \Delta\text{U}^{238}}{\Delta\text{Th} + \Delta\text{U}^{238} + \Delta\text{Pa} + \Delta\text{Np} + \Delta\text{U}^{233} + \Delta\text{U}^{235} + \Delta\text{Pu}^{239} + \Delta\text{Pu}^{241}}$$

Δ difference between start-up and end of cycle. (ΔPa, ΔNp will normally be negative).

$$(2)\ \text{cost} = \frac{(\text{WL}_{\text{fiss}} \cdot p_{\text{fiss}} + \text{WL}_{\text{fert}} \cdot p_{\text{fert}} \cdot \text{WL}_{\text{carb}} \cdot p_c) q^{\Delta t/2}}{\text{P}\eta_e \text{l} \Delta t}$$

cost cost per unit electricity produced.
WL_{fiss} fissile material loaded per cycle.
WL_{fert} fertile material loaded per cycle.
WL_{carb} graphite loaded per cycle.
P thermal reactor power.
η_e total efficiency.
l load factor.
Δt cycle time.
q equated factor for compound interest ($q = 1 + i$, i... interest rate).

$$(2a)\ p_{\text{fiss}} = (p'_{\text{Pu}} + \pi) \cdot q^{t_{\text{Pre}}}$$

$$(2b)\ p_{\text{fert}} = (p'_{\text{fert}} + \pi) q^{t_{\text{Pre}}}$$

$$(2c)\ p_c = p_{\text{element}} \cdot q^{t_{\text{Pre}}}$$

p'_{Pu}; p'_{fert} material price (as applicable).

p_c fuel element cost (graphite component).

π fabrication cost of heavy particles in fuel elements.

t_{Pre} time between date when payment is due and start-up.

ACKNOWLEDGMENT

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APPENDIX A

A BRIEF OUTLINE OF THE BURN-UP PROGRAMME AND NUCLEAR DATA USED

The performance of a plutonium system is very sensitive to the data used. As, however, a thourough investigation of these data is beyond the scope of this report, we shall limit ourselves to the outlining of the basic features necessary for the understanding of the behaviour of the systems.

The Pu^{239} cross sections are relatively well known, especially in the thermal range. The high absorption and fission resonance at 0.3 eV (Fig. A.1) and the important variation of η (Fig. A.2) are of particular interest for the physics of a HTR. The group values of Dragon Library II [2] are in good agreement with the tabulation of J. J. Schmidt [3] under 1 eV. In the resonance range we can more easily compare the resonance integrals above 0.45 eV. We have :

Dragon Library II : $RI_{abs} = 507.1\ b$; $RI_{fiss} = 296.8\ b$; $\eta = 1.71$.

E. Critoph [5] : $RI_{abs} = 516\ b$; $RI_{fiss} = 321\ b$; $\eta = 1.78$ ($\nu = 2.87$).

Some uncertainty seems to remain about the ν-value (2,200 m/g) :

Ref. [2] : $\nu = 2.91$.

Ref. [5] : $\nu = 2.87$.

The other plutonium isotopes, i.e., Pu^{240} and Pu^{241}, are very important owing to their presence in the feed plutonium or to their important production during life (due to the high α-value of Pu^{239} and the high absorption cross section of Pu^{241}). The main feature of Pu^{240} is the giant resonance at 1.057 eV, of parameters :

$\Gamma_\gamma = 0.0325$ eV; $\Gamma_\eta^\circ = 0.00237$ eV.

It produces significant flux dip for usual S-values and the considered grade of plutonium.

The presence and importance of Pu^{241} in the core stems mainly from the high cross section of Pu^{240}. Pu^{241} contributes significantly to the total number of fissions during the core life; unfortunately large uncertainties remain as to its cross sections

and η-value and this may effect markedly the accuracy of the predicted performance as it has been shown for water moderated systems [6]. Fig. A.3 compares the group cross sections used with the Aldermaston compilation [8] (in which η is approximately 2.2 and α 0.4 below 1 eV). The resonance at 0.25 eV is smaller than the neighbouring Pu^{239} resonance and is shielded by it at the beginning of life; at the end) however, when Pu^{241} is important, no shielding (either self or mutual shielding, generally occurs because of the low concentrations. The data above 0.45 eV are [2] :

$$RI_{abs} = 934.4\,b;\quad RI_{fis} = 535.6\,b;\quad \eta = 1.75.$$

The plutonium data given above explains the behavior of the one-group cross section and η-value (Table A.1).

For the present calculations, the burn-up programme HELIOS [1] was used. It evaluates the neutron spectrum in 43 groups at the beginning of life, after Xe and Sm equilibrium poisoning (taken as one day) and at the end of every large timestep. The one group quantities are averaged over the critical spectrum (criticality is achieved by the simulation of control rods by a control poison) and kept constant during the large timestep. The depletion of all isotopes is evaluated by approximating the analytical solution of the burn-up equations over small and variable timesteps.

The group structure of Dragon Library II is too coarse around 1 eV to account correctly for the self-shielding of the Pu^{240} resonance; this means that the group cross sections are in fact dependent on the concentration of Pu^{240} relative to graphite. Fortunately, at usual working conditions, the dependence is small (although not negligible and altering k_{eff} by a few per cent) and the effective cross sections can

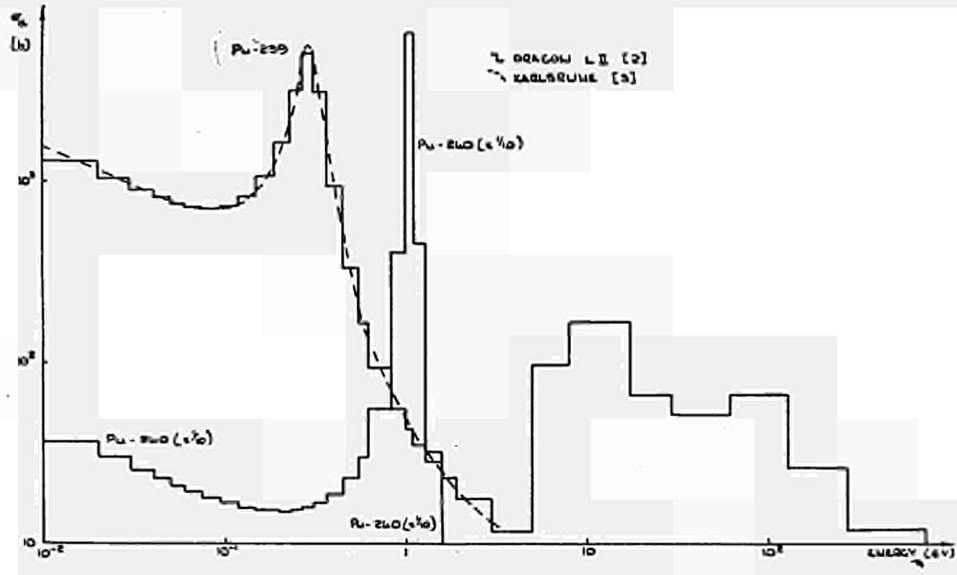

FIG. A.1. — Absorption cross section of Pu^{239}.

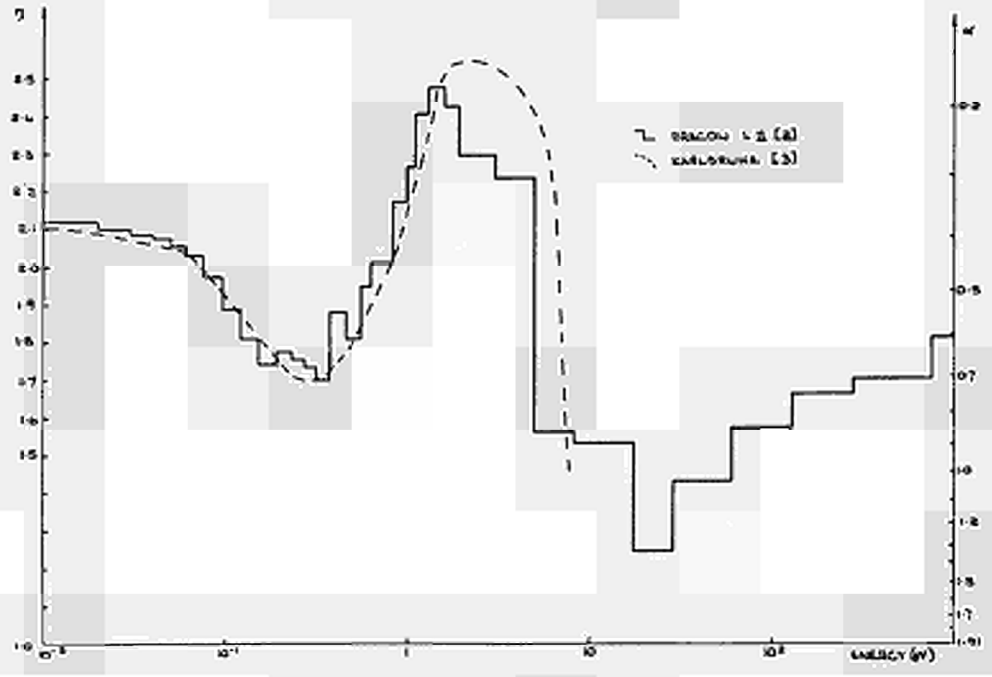

FIG. A.2. — η and α for Pu^{239}.

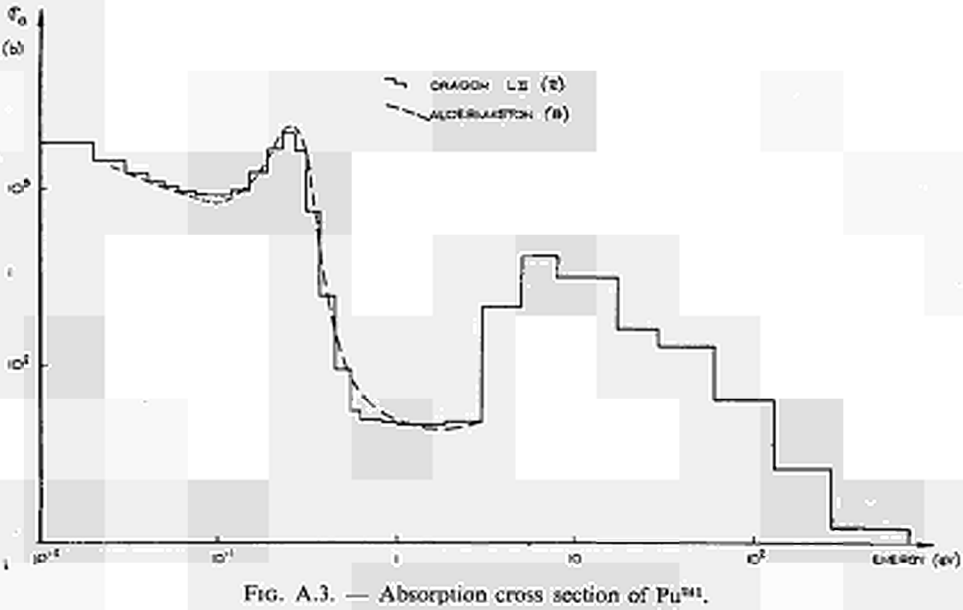

FIG. A.3. — Absorption cross section of Pu^{241}.

TABLE A.1. — Variation of Cross Sections with S and Moderator Temperature

T_{mod}	1300° K		900° K	
S	4,000	10,000	4,000	10,000
$\sigma_a^{Pu\text{-}239}$	104.3	235.3	104.1	231.6
$\eta_{Pu\text{-}239}$	1.760	1.777	1.768	1.794
$\sigma_a^{Pu\text{-}240}$	272.3	356.7	259.8	332.0
$\sigma_a^{Pu\text{-}241}$	95.2	180.0	98.9	196.0
$\eta_{Pu\text{-}241}$	1.937	2.046	1.943	2.048
$\frac{\sigma_a^{Pu\text{-}239}}{\sigma_a^{Pu\text{-}240}}$	0.151	0.257	0.157	0.267
$\sigma_a^{U\text{-}235}$	35.23	55.0	36.7	61.4
$\frac{\sigma_a^{Pu\text{-}239}}{\sigma_a^{U\text{-}235}}$	2.96	4.28	2.84	3.77

be evaluated without working out the most rigorous solutions of the flux. The approximation used is a version of the NRIA (i.e. Narrow Resonance, Infinite Mass) approximation which takes into account the incomplete recovery of the flux below resonance energies and preserves the first and second moments of the Doppler-broadened transfer cross sections. Mathematically the flux is described by a first order differential equation of the Greuling-Goertzel type.

The effective cross sections are inserted in HELIOS by way of an option which allows the cross section of one or more isotopes to be multiplied by disadvantage factors. As only the room temperature Pu^{240} cross sections are written on the library tape, the disadvantage factors must also account for the Doppler-broadening of the resonance. They are constant during life.

We have taken the opportunity of the disadvantage factors option to study the possible effect of spatial self-shielding on the physics and performance of plutonium systems. In order to relate these factors to physical quantities without doing lengthy calculations on complicated fuel elements, we have considered the disadvantage factors of small particles imbedded in a graphite matrix. From the analysis of Lane, *et al.*, on such systems [7], we can deduce the disadvantage factor d, under the assumption of no thermalisation inside the particles :

$$d = \frac{P_o(\Sigma_t r)}{\left(1 - \frac{\Sigma_s}{\Sigma_t}(1 - P_o(\Sigma_t r)\right)}$$

Σ_s, Σ_t scattering, respective total, macroscopic cross section of the kernel material.

P_o escape probability out of the kernel.

r radius of the kernel assumed spherical.

For all thermal groups, except the three spanning the Pu^{240} resonance, d is evaluated using group quantities. For the other three groups d is averaged using the same approximation as for homogeneous systems.

As the disadvantage factors are kept constant during life in the present version of HELIOS, the study of "heterogeneous" systems has mainly an indicative value. Another version, obviating this inconvenience, is being tested.

A second shortcoming of HELIOS is the fact that in plutonium systems, the large timestep between spectrum calculations is to be fairly small to achieve good accuracy, due to the rapid variation of cross sections (Table A.1). Other modifications are under study to circumvent this.

FUEL CYCLES FOR HIGH-TEMPERATURE GAS-COOLED REACTORS UTILIZING PLUTONIUM(1)

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ABSTRACT

Calculations were made of burnup, conversion ratio, and fuel-cycle cost for high-temperature gas-cooled reactors fueled with plutonium-thorium and moderated by graphite. Recycle of the uranium and plutonium, recycle of the uranium only, and non-recycle were considered. The highest conversion ratios (e.g., 0.9 at a burnup of 50,000 MWd/T) were obtained with recycle of the uranium only. An assumed plutonium cost of $ 10.00/g fissile gave minimum fuel-cycle costs of 0. 8mills/kWhr(e) both for the non-recycled fuel and for the recycle of the uranium only. The fuel-cycle costs would be lower than those from uranium-thorium fuels at any plutonium cost below $ 11.00/g fissile.

There has been increasing interest recently in the use of plutonium for fueling thermal reactors. This interest comes mainly not from any superiority of plutonium as a reactor fuel, but form the recognition that plutonium production is increasing in the civilian reactor programs of several countries, providing a source of fuel which must be utilized. The spectrum-averaged η for Pu^{239} is considerably lower, in a typical case, than the η for either U^{233} or U^{235}. For this reason it is likely to prove desirable to use the plutonium to give the needed initial reactivity in the fuel while obtaining a considerable fraction of the fissions in U^{233} or U^{235}. We have studied plutonium-thorium systems in which the conversion of thorium to U^{233} provides fissionable material, either for sale or for recycle to the reactor. The calculations we have done have been for essentially homogeneous mixtures of fuel and graphite with equilibrium, graded-exposure fuel management. Our numerical procedures are described in more detail in Ref. 1.

The feed material in the non-recycle calculations consisted of Pu^{239}-Pu^{240}-Pu^{241}-Pu^{242} in the ratio 52.0 : 29.5 : 11.5 : 7.0, corresponding to the discharge from a pressurized water reactor at 15,000-20,000 MWd/T. This material was also used as makeup in the recycle calculations, along with the material recycled from the reactor in question. Two types of recycle were considered : recycle of all uranium plus plutonium and recycle of only the uranium with sale of the plutonium. In both

(1) ORNL-report TM-1111, April 1965.

(2) Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

types of recycle the recycled material had the asymptotic composition which would be achieved after many cycles. Fixed parameters for the study are given in Table I. Fuel fabrication and reprocessing unit costs were assumed to vary with throughput and were computed from the data in Ref. 1. Remote fabrication was assumed throughout.

TABLE I. — Fixed Parameters for Plutonium Fuel Study

Power density, w/cm³	5.0
Thermal efficiency, %	40
Reactor plant factor	0.8
Average moderator temperature, °K	900
Buckling, cm^{-2}	2.0×10^{-5}
Coolant fraction	0.39
Fixed charges on fuel inventory, % year	10
Fixed charges on working capital, % year	10
Processing losses, % :	
Uranium and plutonium	1.0
Protactinium	3.1
Fabrication scrap losses, %	0.2
Fuel shipping charges, $/kg of heavy metal :	
To processing plant	6.40
From processing plant	5.05
Cost of plutonium, $/g fissile	10.00
Cost of fissile uranium, $/g [a]	12.05

[a] This price is for fully enriched material. The USAEC cost schedule was used for lower enrichments.

The principal variables in the study were the type of fuel management and the composition of the fuel. The burnup and conversion ratio that were obtained are shown in Figs. 1 through 3 for the three types of fuel management as a function of composition. Although it is not evident from the graphs, each type of fuel has its maximum conversion ratio when the initial moderator-to-fissile ratio is 5,000-6,000. The highest conversion ratios (0.9 or above) were obtained with the recycle of the uranium but not the plutonium. In this method of fuel management full advantage is taken of the bred U^{233} by the recycle of the uranium, but the Pu^{242} is not allowed to build up to levels at which it becomes a significant poison. In the optimum case the concentration of fissile plutonium is reduced to less than 1 % of its initial value in a single cycle, so the plutonium could be discarded at this point without any economic penalty.

Non-recycle plutonium-thorium fuel gives conversion ratios that are somewhat lower (a maximum of about 0.8). However, the uranium available for sale from this type of fuel management has a composition that would be desirable for any thermal reactor. In the optimum case the $U^{233} : U^{234} : U^{235} : U^{236}$ ratios in the discharged fuel are 88.9 : 9.7 : 1.3 : 0.1.

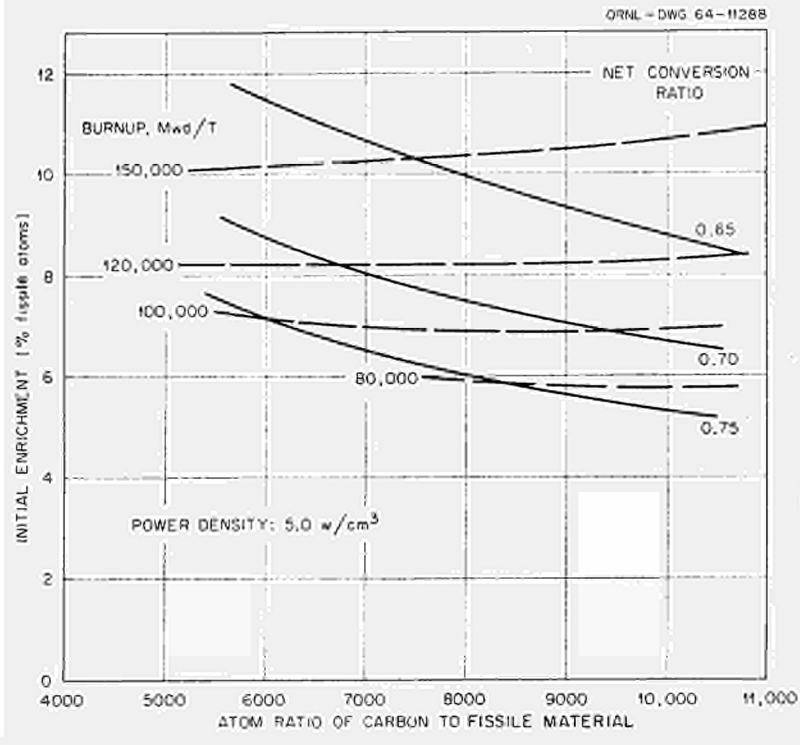

FIG. 1. — Burnup and Conversion Ratios for Non-Recycled Plutonium-Thorium Fuel.

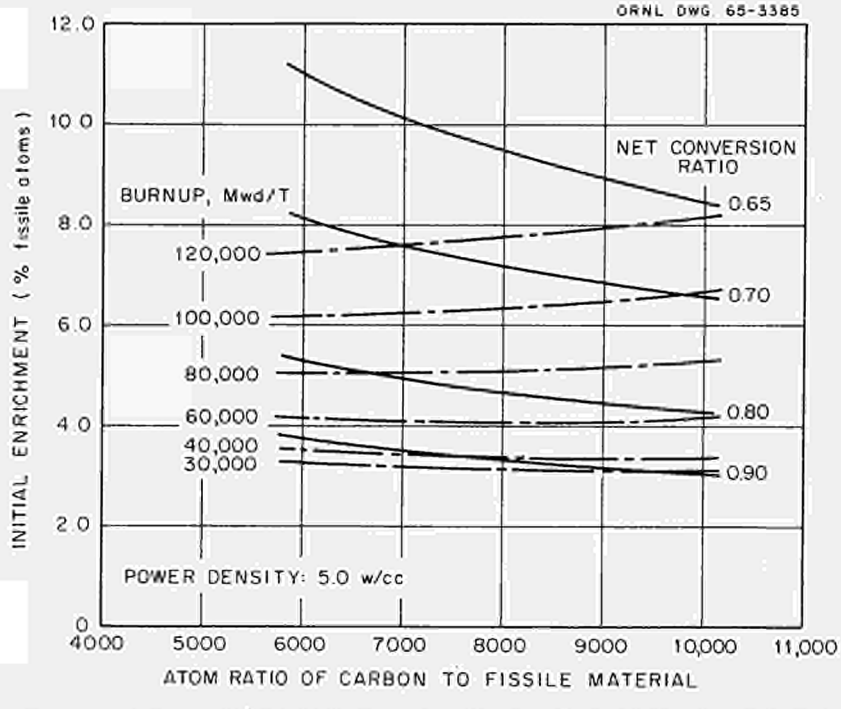

FIG. 2. — Burnup and Conversion Ratios for Plutonium-Thorium Fuel with Uranium Only Recycled.

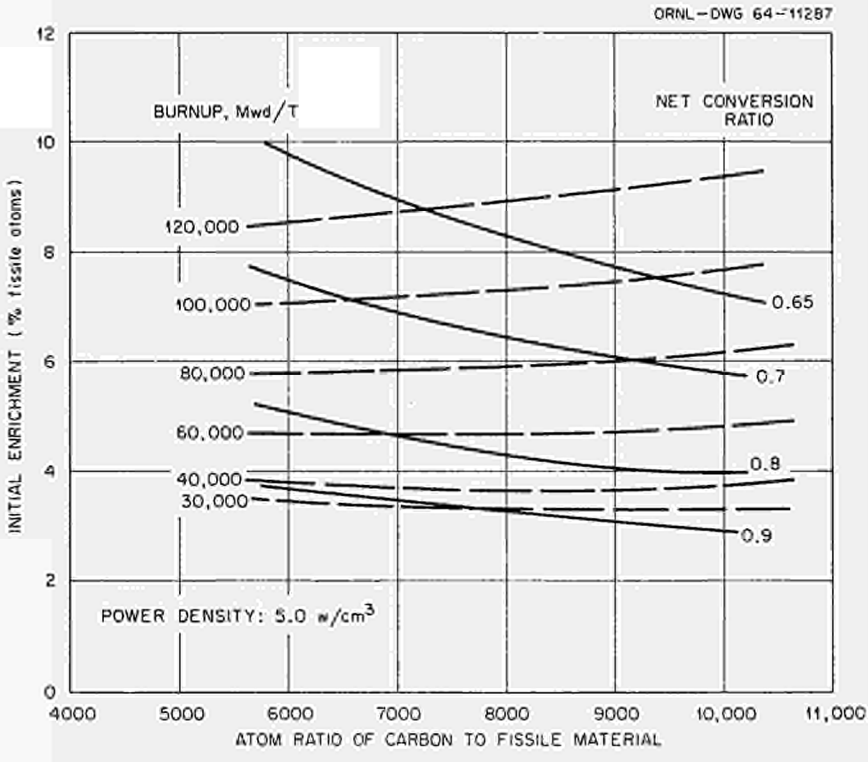

FIG. 3. — Burnup and Conversion Ratios for Plutonium-Thorium Fuel with Uranium and Plutonium Recycled.

TABLE II. — Minimum Fuel-Cycle Cost for Plutonium Fuels

	Minimum Fuel-Cycle Cost, [mills/kWhr(e)]		
	Not Recycled	Uranium Only Recycled	Uranium and Plutonium Recycled
Processing	0.163	0.158	0.164
Fabrication	0.146	0.114	0.128
Fabrication interest	0.013	0.007	0.011
Shipping	0.008	0.012	0.015
Plutonium feed	0.809	0.377	0.446
Uranium credit	(0.396)		
Plutonium credit	(0.060)	(0.001)	
Core inventory	0.070	0.097	0.081
Fabrication inventory	0.029	0.030	0.037
Processing inventory	0.021	0.014	0.018
TOTAL	0.803	0.808	0.900
Total with spent fuel discarded[a]	0.931		

[a] Fuel composition was reoptimized in computing the costs for this line.

Full recycle of uranium and plutonium gives conversion ratios that are lower than for the recycle of the uranium only and does not appear to give any compensating advantage. As mentioned above, the fissile content of the recycled plutonium would be low, and the advantage of using Pu^{240} as a fertile material is more than offset by the continued buildup of Pu^{242}. We have not investigated the possibility of recycling the plutonium for just a few cycles. Such a procedure could be worthwhile for a low-burnup reactor.

The fuel-cycle costs for the optimum (economic) case in each of the three types of fuel management are detailed in Table II. It can be seen that the non-recycled fuel and the fuel in which the uranium only is recycled give approximately equal total costs of 0.8 mills/kWhr(e). The full recycle of uranium and plutonium gives a cost 0.1 mills/kWhr(e) higher. It is also to be noted that the discharged fuel from the non-recycle case could be discarded or stored without being reprocessed instead of being sold, giving a cost of 0.9 mills/kWhr(e).

Fuel compositions and other data for these same cases are given in Table III. The isotopic neutron balances are indicated in Table IV. The initial moderator-

TABLE III. — Plutonium-Thorium Compositions Yielding Minimum Cost

	Not Recycled	Uranium Only Recycled	Uranium and Plutonium Recycled
Fresh fuel composition :			
Moderator-to-fissile atom ratio. .	10,000	8,000	10,000
Enrichment, % fissile atoms . .	5.38	6.03	5.90
Average core composition :			
Moderator-to-fissile atom ratio .	18,400	15,900	18,400
Enrichment, % fissile atoms . .	3.10	3.26	3.36
Reactivity lifetime :			
Cycle time, full power days . . .	519	771	503
Fissions per initial fissionable atom	1.38	1.64	1.33
MWd/T (U + Pu + Th). . . .	72,000	95,000	76,000
Average core specific power, kw/kg fissile	4,610	4,030	4,730
Net conversion ratio	0.75	0.74	0.70
Plant throughput, MT/year :			
Processing	139	103	137
Fabrication	153	115	136
Unit cost for fabrication plus processing, $/kg	224	266	225
Ratio of initial to average power density	2.38	3.22	2.79
Average η of fissile nuclides	2.04	2.13	2.12

TABLE IV. — Neutron Balance for Minimum Cost Plutonium-Thorium Fuels

	Reactions Per Source Neutron		
	Not Recycled	Uranium Only Recycled	Uranium and Plutonium Recycled
Absorptions :			
Th^{232}	0.185	0.228	0.197
Pa^{233}	0.006	0.007	0.006
U^{233}	0.075	0.220	0.189
U^{234}	0.002	0.028	0.024
U^{235}	0.001	0.028	0.024
U^{236}	0.000	0.005	0.004
Np^{237}	0.000	0.002	0.001
Pu^{239}	0.202	0.101	0.118
Pu^{240}	0.186	0.098	0.114
Pu^{241}	0.211	0.118	0.139
Pu^{242}	0.025	0.020	0.052
Fission products	0.068	0.094	0.077
Moderator	0.019	0.028	0.031
Leakage	0.020	0.023	0.024
TOTAL	1.000	1.000	1.000
Productions :			
U^{233}	0.168	0.497	0.428
U^{235}	0.001	0.056	0.048
Pu^{239}	0.354	0.178	0.208
Pu^{241}	0.476	0.266	0.313
Other	0.001	0.003	0.003
TOTAL	1.000	1.000	1.000

to-fissile ratios for the economic optimum cases (8,000 to 10,000) are substantially higher than those for maximum conversion ratio, principally because of the lower fuel inventory cost associated with higher specific power.

The fuel-cycle costs for these plutonium-thorium fuels can also be compared with those for the uranium-thorium and partially enriched uranium fuels [1], but it must be kept in mind that the comparison is extremely sensitive to the assumed value of plutonium. At the $10.00/g fissile assumed in this study the best cost for a plutonium-thorium fuel was 0.80 mills/kWh(e). Under the same ground rules the best cost for a uranium-thorium fuel was 0.88 mills/kWhr(e) and the best cost for a partially enriched uranium fuel was 0.93 mills/kWhr(e). In another way of

looking at the same question, the plutonium-thorium fuel appears to compete favorably with the uranium-thorium fuel as long as the price of plutonium is less than $11.00/g fissile.

Two disadvantages relating to plutonium-thorium fuels in high-temperature gas-cooled reactors have not yet been fully analyzed. The first is that the relatively high fission cross section of the plutonium isotopes compared to U^{233} results in a large decrease in power for a fuel element as the plutonium burns out. The ratio of initial-to-average power from this factor, as given in Table III, would be 2.4 to 3.2 for the various types of fuel management. The power peaking could be reduced by lower burnup of the fuel, but only at an appreciable penalty in fuel-cycle cost. In order to utilize effectively the heat removal capacity in the core it might prove necessary to use two different fuel elements with different fuel compositions and different core residence times. The other persistent question which has apparently not yet been completely answered is whether fuels with a high plutonium content would have an unsatisfactory temperature coefficient.

If favorable answers to these two questions can be obtained, it appears that the high-temperature gas-cooled reactor is well suited to utilizing available plutonium supplies. The fuel-cycle costs in a plutonium-thorium cycle would be at least as attractive as those in other cycles under the most probable projection of economic assumptions.

REFERENCE

1. R. S. CARLSMITH, C. M. PODEWELTZ and W. E. THOMAS. — *Fuel-Cycle Cost Comparisons for High-Temperature Gas-Cooled Reactor Fuels.* This Symposium, p. 357.

A REVIEW OF THE STUDIES MADE ON THE UTILISATION OF PLUTONIUM IN THE HIGH TEMPERATURE REACTOR

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ABSTRACT

The paper discusses the reactor physics studies carried out during 1961/1962 at A. E. E. Winfrith on the utilisation of plutonium in the High Temperature Reactor. The calculational model is described with particular reference to the Pu^{240} isotope. The paper considers temperature coefficients, long term reactivity changes and fuel cycle aspects of the design.

1. — INTRODUCTION

1.1. The purpose of this paper is to discuss the reactor physics studies carried out during 1961/1962 at A. E. E., Winfrith on the utilisation of plutonium in the High Temperature Reactor. The reactor model used for the assessment was a 1,000 Mw heat output system, graphite moderated and based essentially on the DRAGON concept.

1.2. Previous studies of the fuel cycle performance of the HTR fuelled with U^{235} as fissile material and Th^{232} as the fertile have indicated the need for large supplies of U^{225} from a diffusion plant if a power programme of HTR's is to be supported. In view of the fact that in the future the Magnox power programme in the UK will be producing large quantities of civil plutonium, it was decide to examine this material as the feed instead of U^{235}. The plutonium used in the calculations has isotopic concentrations consistent with 3,000 MWD/teU irradiation in a civil power station, and these are taken to be Pu^{239} 80 %, Pu^{240} 16 % and Pu^{241} 4 %.

2. — REACTOR PHYSICS

2.1. — CALCULATIONAL METHODS.

2.1.1. In the designs of HTR at present contemplated the fuel is mixed with a large amount of moderating material to form a near homogeneous core. This means that, from the reactor physics point of view, the neutron flux is flat across the cell and the reactor may be considered to be homogeneous for all reactions except those in resonances; this represents the simplest of calculational problems from the flux fine structure point of view.

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(1) UKAEA, Reactor Group, Technical Assessments and Services Division. Atomic Energy Establishment Winfrith, Dorset, Great Britain, Report W.6945 A, April 1965.

2.1.2. The STEWPOT Code is designed to study the reactor physics characteristics of HTR cores and in its basic form considers the medium to be completely homogeneous except for resonance captures. The calculational procedure is to compute the neutron energy spectrum from fission energies to thermal by means of a 43 energy group representation. The 20 groups below 1 eV may be considered as thermal and the representation of the thermalisation processes is by either the free gas or the Schofield crystal scattering law. The resulting neutron spectrum is then used to condense the 43 group data to few groups. These few group data are then used for reactivity burn-up and temperature coefficient calculations.

2.1.3. From the initial calculations on plutonium fuelling, it became clear that the plutonium-240 component in the civil plutonium was to play an important role in the fuel cycle performance of the system, since it was shown that the neutron captures could account for 40 % in reactivity. This effect is a result of the under-moderation of the HTR, the homogeneous form of the core and the slowing down characteristics of graphite. The neutron captures predominate in the broad resonance exhibited by Pu^{240} at 1 eV and because of this considerable effort has been devoted to the establishing of methods for assessing the reaction rate in this resonance with particular attention being paid to fuel element heterogeneity.

2.1.4. There are two basic problems associated with the evaluation of the resonance escape probability which prevent the use of standard and proven techniques used in the cases of U^{238} and Th^{232}. These are :

(i) the resonance is situated at the lower energy end of the slowing down region and, at 1 eV, the upscatter of neutrons by thermalisation processes to energies above the resonance becomes significant. This is particularly so in the HTR with neutron temperatures of 1,100° K. This means that a neutron faces the probability of capture during the slowing down stage and having escaped capture may subsequently be raised in energy by means of a collision with a moderator atom to a level above the resonance, thereby increasing the probability of capture.

(ii) a simplifying assumption frequently used in the evaluation of resonance capture is the method known as the Narrow Resonance Approximation (NRA); this is based on the approximation that the resonance width is small compared to the energy loss a neutron suffers in a collision with the moderator. In the case of Pu^{240} with graphite or beryllia this assumption is inadmissible since the resonance width is large compared to the maximum energy loss, and because of this the neutron spectrum on the lower energy side of the resonance is suppressed due to neutron capture at the higher end. The use of the NRA in this case does lead to an overestimate of neutron captures particularly when the plutonium concentration is high.

2.1.5. The method of calculation for determining the self-shielding is based on multigroup neutron transport theory. The energy range in the region of the resonance is subdivided into many small groups and the precise number required

to provide consistent results is determined by trial and error. A source of neutrons is assumed to be injected into the reactor at an energy well above the resonance and the subsequent slowing down of these neutrons from group to group through the resonance is computed. The resonance escape probability may be evaluated by determining the number of neutrons remaining below the resonance as a fraction of the starters. Allowance is made for neutron upscatter by including a transfer matrix of scattering cross-sections from energies below the resonance to higher energy. The use of this method, which is embraced in a computer programme, allows cross-sections to be tabulated for various geometries in the wider group form required for STEWPOT.

2.2. — Experimental Verification of the Theoretical Methods.

2.2.1. The STEWPOT method of calculation for HTR has been used extensively by the DRAGON project for the analysis of HTR cores fuelled with Uranium 235. This experience is relevant to plutonium utilisation since it serves to check the calculation systems together with such things as the basic scattering data for graphite which are common to both plutonium and uranium fuelling.

2.2.2. STEWPOT has also been used for the analysis of a series of plutonium graphite exponential experiments in the SCORPIO facility. This experiment unfortunately was constructed of plutonium bearing spikes with only 2.5 % plutonium 240 content (instead of the 16 % in civil plutonium). The agreement reached between theory and experiment was reasonable, with a maximum error of 2 % in reactivity, but the low plutonium-240 concentration limits the value of this experimental check of the theory.

2.3. — Temperature Coefficients.

2.3.1. The temperature coefficient of reactivity associated with the moderator of the HTR with plutonium fuelling remains negative over a wide range of conditions.

2.3.2. The moderator temperature coefficient has components due to :

(i) changes in thermal utilisation (f)

(ii) changes in the neutron yield (η)

(iii) changes in thermal neutron leakage (L^2).

The change in thermal utilisation results from the relative captures in fissile material and the capture in other materials. As the moderator temperature is increased, the neutron energy spectrum moves into the Pu^{239} resonance at 0.3 eV and the effective thermal absorption cross-section remains nearly constant; the effective cross-sections of the other reactions fall since these are in general "1/v". The relative captures therefore in the Pu^{239} increase with temperature and this leads to an increase in thermal utilisation. Since the HTR has a thermal utilisation factor approaching unity the positive component to the overall coefficient is small. It should be noted, however, that should some additional parasitic absorber be added to the design then this will lead to an increased positive component to the

coefficient. The change in the neutron yield (η) results simply from the decreasing value of this parameter as the neutron energy is increased, and this component is negative. The change in the leakage is small since the absorption cross-section of Pu^{239}, already mentioned, remains nearly constant, as does the scattering cross-section.

2.3.3. The change in eta with temperature is the largest component and is negative and off-sets the small positive component from the thermal utilisation. There are, however, indications that as the burn-up of the reactor proceeds, the temperarature coefficient becomes less negative and at the end of core life may even be slightly positive. This is the result of increased parasitic absorption from the fission product poisons. The changing temperature coefficient would require examination from the operational point of view and could limit the lifetime of core charge.

2.4. — Long Term Reactivity Changes.

2.4.1. The reaction chain relevant to plutonium burning is :

$$Pu^{239} \longrightarrow Pu^{240} \longrightarrow Pu^{241} \longrightarrow Pu^{242}$$

Radiative capture in plutonium leads to the production of Pu^{240} and captures in this isotope lead to the production of Pu^{241}, which is fissile. The high neutron losses in Pu^{240} can be considered therefore as a component to the conversion factor of the system. It is of interest to note also that the eta value of the Pu^{241} produced is higher than that of Pu^{239} and is in fact greater than that of U^{233} in these spectrum conditions. Typical values for the four fissile materials are given in Table 1.

Table 1. — Eta values for the various fissile isotopes

U^{233}	U^{235}	Pu^{239}	Pu^{241}
2.12	1.74	1.75	2.18

As operation proceeds the Pu^{239} content burns up whereas the Pu^{241} and the U^{233} from Th^{232} will increase. It is to be expected therefore that the value of eta averaged over all materials in the core will increase and this will lead to a release in reactivity which tends to compensate for other losses.

2.4.2. For a completely homogeneous core, the burn-up rate of Pu^{240} is considerably higher than the production rate from Pu^{239}. This means that the concentration of Pu^{240} and also the reactivity absorbed by this isotope falls with time in a similar manner to a burnable poison; this leads to a steady release in reactivity with time which more than off-sets the negative changes in reactivity. The burn-up of Pu^{240} and the resulting release in reactivity may be adjusted by self-shielding the resonance. By a suitable choice of fuel element configuration, a flat reactivity/time curve may be achieved. Examples of this will be given in the next section of the paper.

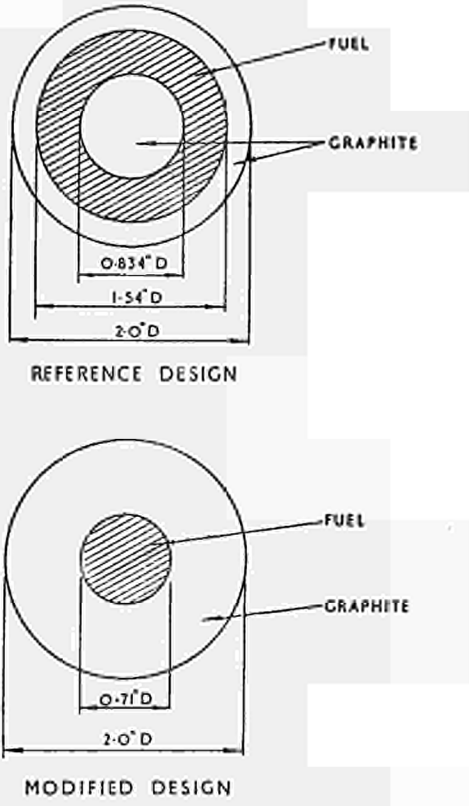

FIG. 1. — Fuel Element Configurations.

2.5. — FUEL CYCLE PERFORMANCE.

2.5.1. The reactor chosen as a model for the fuel cycle study has a 1,000 MW(Th) output and is based on the DRAGON reactor concept. The fuel element selected for study is shown in Figure 1 and consists of an outer graphite sleeve which covers a fuel insert surrounding a central graphite spine. In practice the outer surface of

TABLE 2. — Parameters of the HTR model

Item	Power Output
Thermal Output (MW)	1,000
Plutonium Loading	1,000-667 kg[a]
Core Diameter (ft)	16
Core Length (ft)	16
End Reflector Thickness (ft)	1.64
Core Voidage (%)	24
Graphite/Pu (Total) Atom ratio	2,621-3,946
Thorium/Pu (Total) Atom ratio	2.2-7.2
Graphite/Pu Atom ratio in fuel insert	1,102-1,653
Atom/cc of Pu in fuel insert ($\times 10^{-20}$)	0.819-0.546

[a] A series of core conditions were examined and the figures given here indicate the range of interest.

the sleeve is hexagonal in form but has been cylindricalised for convenience of computation. Also in Figure 1 is shown a proposed modified fuel arrangement; a comparison of the fuel cycle performance of the two fuel elements will be made.

2.5.2. The parameters for the reactor are given in Table 2.

2.5.3. The dimensions of the core and therefore the power density are fixed from mechanical design considerations; the plutonium loading is selected to give the required core lifetime which is estimated essentially from economic arguments. An important difference between this reactor and one fuelled with uranium-235 is the level of thorium. In the latter case it may be expected to have for each fissile atom between 10 and 15 thorium atoms whereas in the case of plutonium the thorium atoms may be between 2 and 7. This higher concentration of fissile material in the heavy atoms may well have material problems associated with it.

2.5.4. In Figure 2, the reactivity variation as a function of time for the reference core configuration is shown. It will be noted that the reactivity release rate is excessive and rises to an excess value of 17.5 %; this is a result of the fact that the Pu^{240} is being destroyed too rapidly. The effect of modifying the fuel arrangement is illustrated and it will be noted that the reactivity/time curve is vastly improved; this result comes from tailoring the rate of burn-up of Pu^{240} and provided suitable fuel

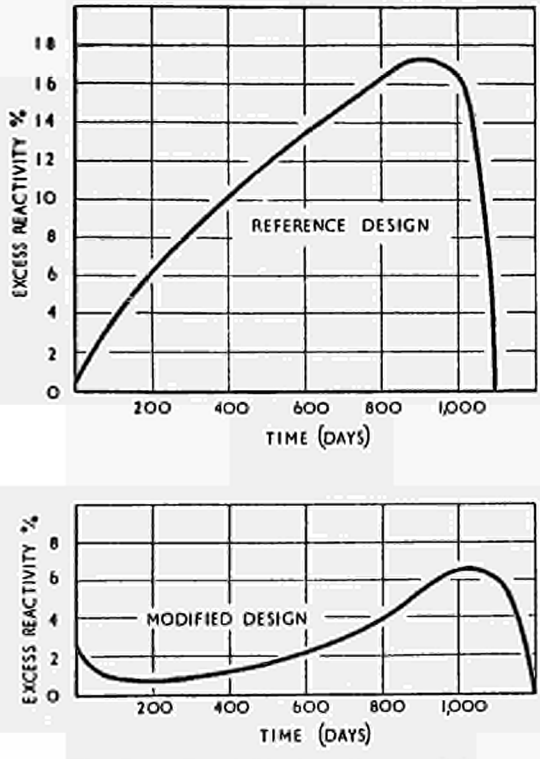

FIG. 2. — Reactivity/Time Characteristics.

designs can be evolved a fuel charge could be taken to high irradiation with little change in reactivity. This means that a complete core replacement method of operation could be envisaged, eliminating the complex operation of zone refuelling required with U^{235} fuel.

2.5.5. From the physics point of view, high burn-up of the initial plutonium appears to be possible to such an extent that 80 % is fissioned. For the range of designs studied the fissions/cc of insert lie in the range 0.5 to 0.75 $\times$ 10^{20} for the reference fuel element and 1.5 $\times$ 10^{20} to 2.25 $\times$ 10^{20} for the modified version. The required increased irradiation performance of the modified fuel element would have to be set against the physics and operational advantages previously described.

2.5.6. The U^{233} produced from Th^{232} was recycled in later fuel charges and the calculations indicate that this will build up to a significant level. In Figure 3, the change in the initial fissile content of successive loads is demonstrated and it will be seen at the end of reactor life 50 % of the fuel is U^{233}. This procedure is not necessarily the optimum method of operation and it may be better to extract theU^{233} from the reject fuel for utilisation in a reactor system operating on the U^{233}-thorium fuel cycle.

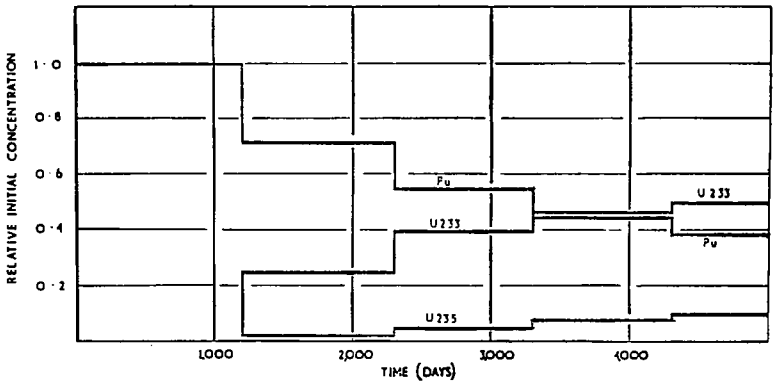

FIG. 3. — Variation of fissile Content at the Beginning of successive Fuel Charges.

2.6. — ALTERNATIVE FUEL CYCLES WITH PLUTONIUM FUEL.

2.6.1. As an alternative to the fuel cycle described, the Pu^{239}-U^{238} cycle has been briefly considered. The physics performance was found to be only slightly different to the Pu^{239}-Th^{232}; this difference comes from the fact that Pu^{239} is built up as the secondary fissile material instead of U^{233}.

2.6.2. The use of plutonium from the AGR instead of that from Magnox reactors was considered, the point being that the two cases have different isotopic concentrations. It was found that the difference in performance was insignificant.

3. — CONCLUSIONS

(i) Theoretical methods have been developed for studying the utilisation of plutonium in the HTR Some experimental verification of these methods has been

achieved. There remains a need for experimental work with high Pu^{240} content fuel.

(ii) The calculations indicate that by careful choice of fuel arrangement a flat reactivity/time characteristic may be obtained; the possible use of a complete core replacement is to be noted instead of the continuous charge/discharge required with U^{235} fuelling.

(iii) From the physics point of view high burn-up of he fuel is possible with up to 80 % of the initial plutonium undergoing fission.

(iv) The temperature coefficient of the moderator is negative at the start of life and decreases as burn-up proceeds with the possibility of its becoming positive at the end of life.

SOME ASPECTS OF THE USE OF PU IN HIGH-TEMPERATURE REACTORS

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ABSTRACT

The paper consists of two parts.

The first part deals with the physical problems involved in the study of Pu fuel in High Temperature Graphite Reactors and with the methods of calculation used.

The second part deals with the study of batch loading fuel cycles in this type of reactors, considering different moderation ratios and fuel lumpings.

1. — PHYSICAL PROBLEMS INVOLVED IN THE STUDY OF Pu FUEL IN HIGH TEMPERATURE GRAPHITE REACTORS, AND METHODS OF CALCULATION USED

The main characteristics of Pu fuel in HTGR type reactors are due to the high cross sections and very pronounced resonances of Pu^{239} and Pu^{240}.

This fact involves serious problems for spectra calculations. If one wants to stay within the multi-group formalism using microscopic un-adjusted cross sections, it is therefore necessary to use a large number of thermal groups.

Another problem arises from taking into account the heterogeneous cell structure of the reactor.

The high resonance cross section of Pu^{239} and Pu^{240} produces a considerable self-shielding. The flux structure in the resonance energy range is also strongly influenced by the up-scattering so that it is not any more possible to apply the methods used for treating the resonances in the fast energy range.

It seems therefore advisable to go to multi-group DSN calculations to obtain reliable Pu self-shielding factors.

Also the variation of this self-shielding with irradiation is very considerable and it is therefore necessary to take it into account in the burn-up calculations.

Because of all the above mentioned considerations it has been decided to use 96 thermal groups (up to 2.0 eV) and 68 fast groups (using the General Atomic GAM [1] and Gather multigroup libraries) and to repeat the spectra calculations as many times as needed during the reactor life.

The work resulted in the development of the MAFIA II code which couples the multi-group spectra calculation with a one-dimensional 4-group burn up calculation [2].

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(1) C.C.R. Ispra, Research and Training Division, Reactor Physics Department, April 1965.

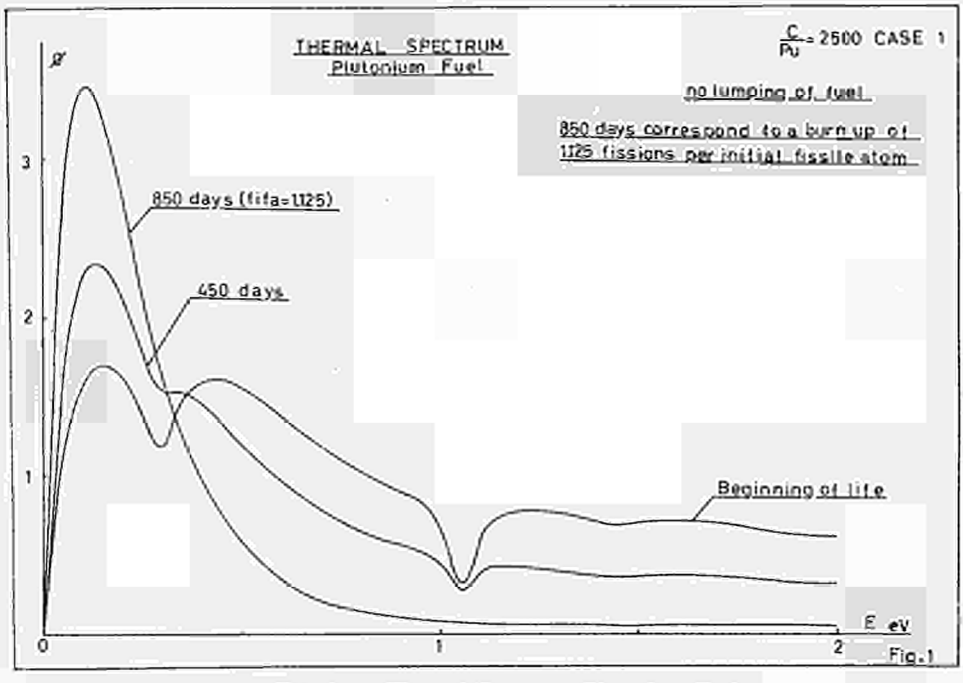

FIG. 1. — Thermal Spectrum, Plutonium Fuel

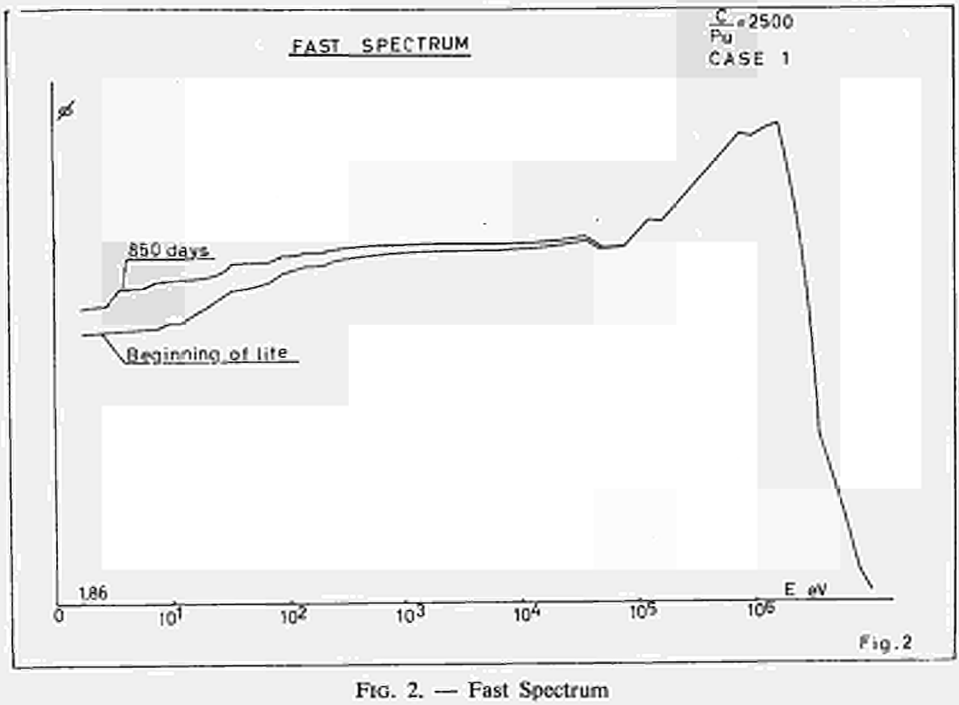

FIG. 2. — Fast Spectrum

The spectrum is used to produce averaged few-groups constants or diffusion and burn-up calculations. The spectrum calculation is zero dimensional with bucklings obtained from the one-dimensional few group calculation. The P1 approximation is used for the fast spectrum calculation, while Po has been considered sufficient for the thermal calculation.

For the self-shielding treatment it was decided to make a few DSN calculations with different Pu concentrations and then to insert a fitting of the self-shielding as function of concentration in the burn-up calculations.

The following formula was used for the fitting.

$$ss = \frac{1}{\sqrt{1 + Ax + Bx^2}}$$

where :

x = concentration.

A and B obtained from a fitting with the DSN results.

These self-shieldings are applied to small energy groups around the Pu^{240} and Pu^{239} resonances, being x the Pu^{240} and the Pu^{239} concentration respectively.

Spectral variations during the reactor life are plotted in Fig. 1-4 for various compositions and fuel lumpings. As a comparison is shown the spectral variation in a U^{235}-Th loaded reactor of the same type (Fig. 5).

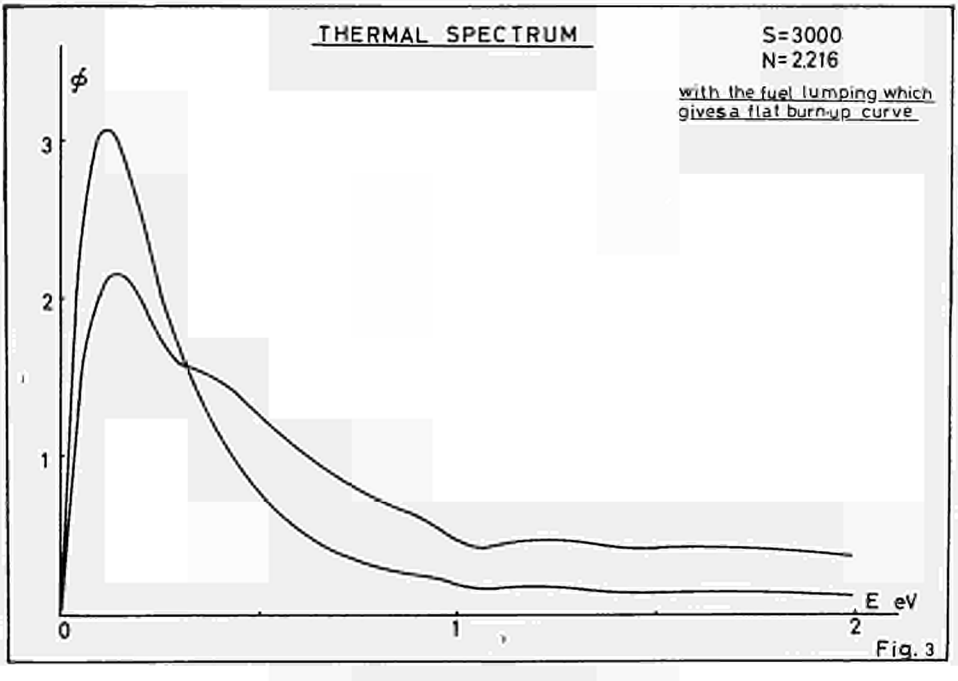

FIG. 3. — Thermal Spectrum

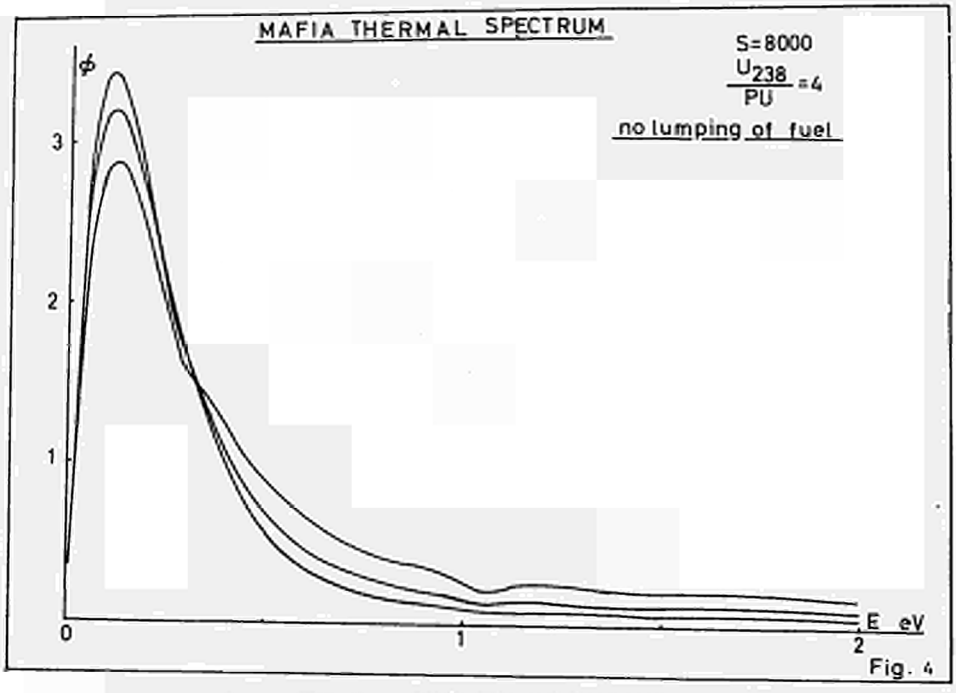

FIG. 4. — Mafia Thermal Spectrum.

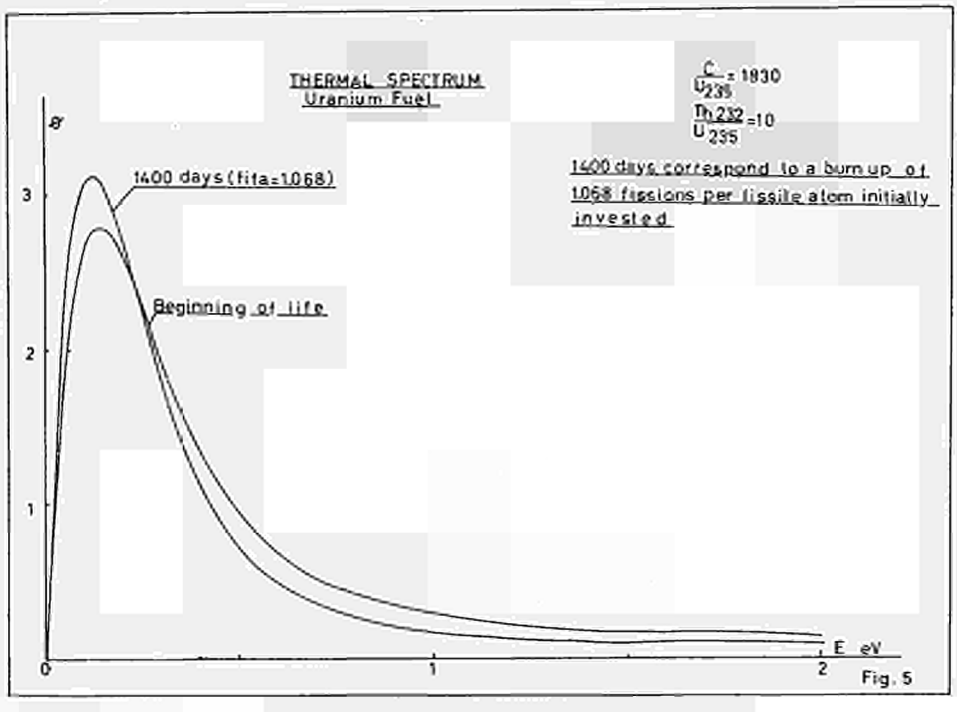

FIG. 5. — Thermal Spectrum, Uranium Fuel.

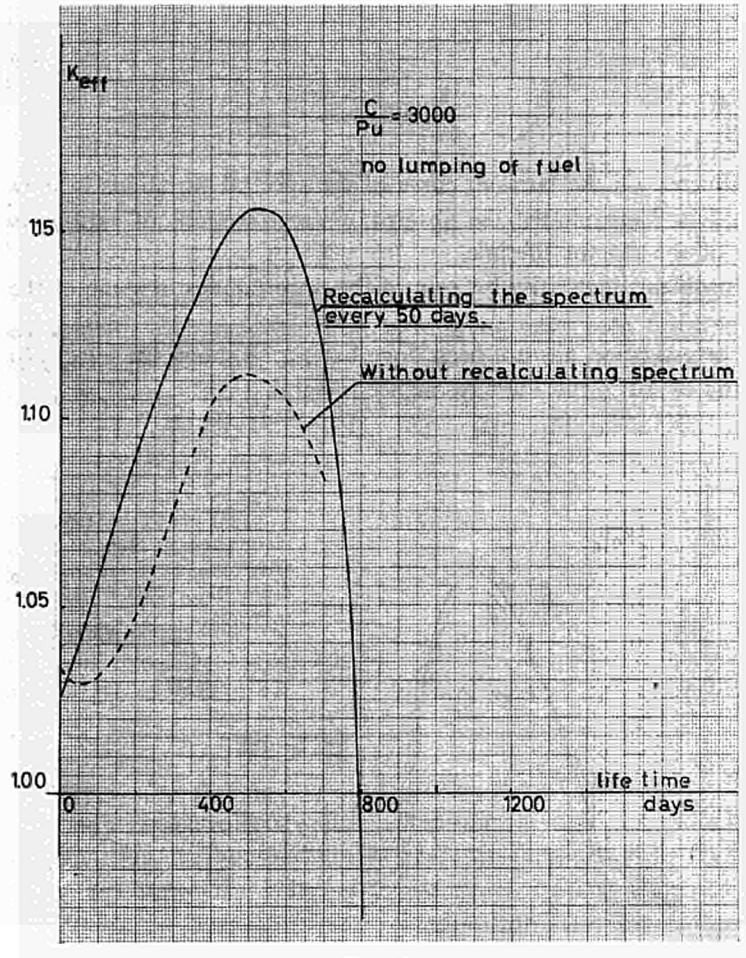
K_{eff}
$\frac{C}{Pu} = 3000$
no lumping of fuel
1.15
1.10
1.05
1.00
Recalculating the spectrum every 50 days
Without recalculating spectrum
0
400
800
1200
life time
days

FIG. 6.

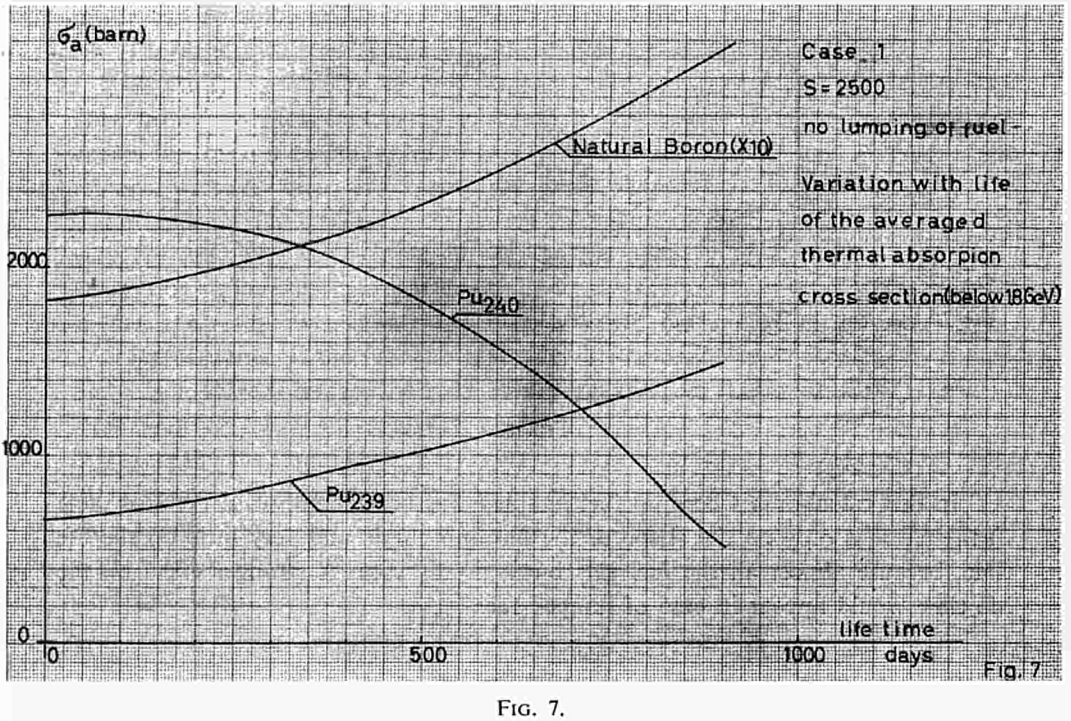
σ_a (barn)
Case_1
S = 2500
no lumping of fuel
Variation with life of the averaged thermal absorpion cross section(below 1.86eV)
Natural Boron(X10)
Pu_{240}
Pu_{239}
2000
1000
0
0
500
1000
life time
days
Fig. 7

FIG. 7.

The influence on the burn-up curve of the spectral variations is shown in Fig. 6, while in Fig. 7 are plotted the absorption cross sections of various isotopes as a function of the reactor lifetime.

Some problems may arise because of the uncertainties in some of the Pu cross sections.

A full investigation has not been done but Fig. 8 shows the effect of increasing or decreasing by 10 % the Pu^{240} resonance integral.

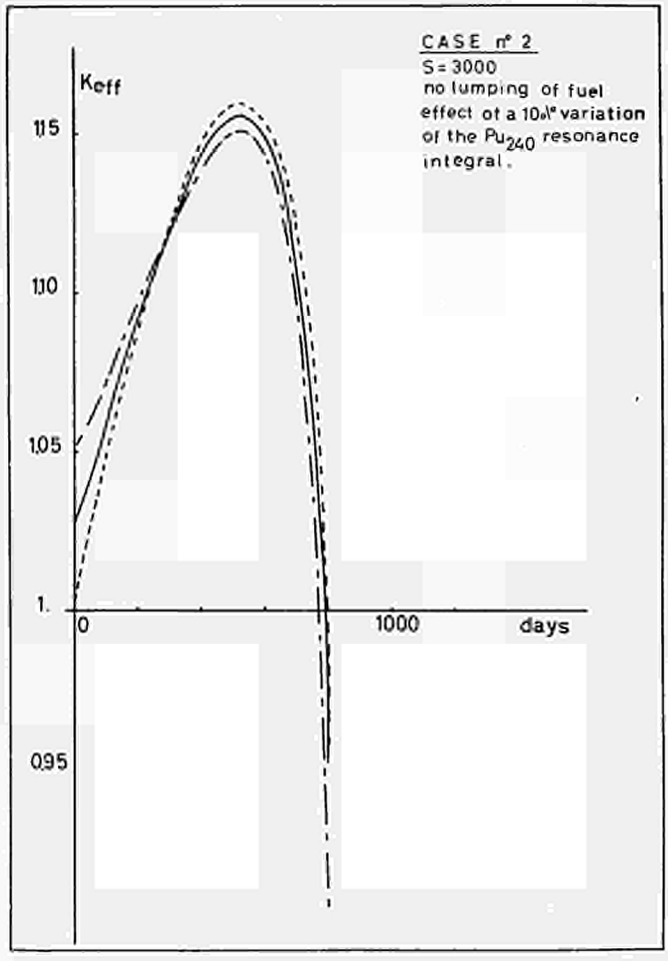

FIG. 8.

In this calculation where no fuel lumping was considered, (case N° 2 of table 2) the effect is only on the shape of the burn-up curve, and very little on the life time, because almost all the fuel gets burnt in any case. A stronger effect on the life time would appear in case of lumped fuel.

2. — Batch Loading Burn-up Studies

Some batch loading studies have been made on Pu fuelled high temperature graphite reactors.

Different initial moderator to fuel (S) and fertile to fissile (N) ratios have been considered, and for each case various fuel lumping have been taken into account.

For these studies the total power was fixed to 1,000 thermal MW and the dimensions are given in table 1.

Table 1

Total thermal power (MW)	1,000
Power density (MW/m^3)	10
Core radius (m)	2.86
Core height (m)	3.89
Fuel average temperature (°K)	1,500
Moderator average temp. (°K)	1,200
Reflector average temp. (°K)	900

The power density has no influence on the physics behaviour of the reactor if no Th is present, provided one can keep the same average moderator temperature.

This means that the consequence of these assumptions is to fix the leakage.

The fuel, moderator and reflector temperatures are also shown on table 1.

The effects on the burn-up of some variation of these temperatures has been checked. This is not very great if the variations are not such as to alter the concept of high temperature reactor. Within these limits a reduction of the moderator temperature can be compared with a slight increase of the S value.

The results of these batch loading studies are presented in table 2.

As the fuel geometry has not yet been fully investigated and only a few cases have been until now calculated in Ispra, it was preferred to quote the burn-up curves in terms of beginning of life self-shieldings in the Pu^{239} and Pu^{240} resonance regions, without connecting it with the actual shape of the fuel element.

It appears that because of the strong conversion of Pu^{240} it is possible to achieve very flat reactivity versus time curves. On the other hand, because of the strong absorption in Pu^{240} it is not possible to insert great amounts of other fertile materials so that the reactivity will suddenly drop after the burning of all the Pu.

The lumping of the fuel can influence this behaviour : with strongly lumped fuel it is possible to reduce the Pu^{240} absorption and then have part of the conversion from another fertile material (Th or U^{238}).

Actually the lumping needed varies with the moderation ratio. In highly moderated systems the flux in the Pu^{240} resonance is rather low, while it is higher in undermoderated systems. This means that with low S values it is necessary to use a more lumped fuel than with higher S values, for obtaining a flat reactivity versus time curve.

TABLE 2

S	N	Pu^{240} Self-shielding (from 0.99 eV to 1.11 eV)	Pu^{239} Self-shielding (from 0.25 eV to 0.35 eV)	Life time (days)	Fifa	Maximum / Average power		Case No
						BOL	EOL	
Pu-Th cases								
2,500	2.5	1.0	1.0	861	1.07	1.546	1.945	1
3,000	3	1.0	1.0	710	1.06	1.5312	1.8065	2
4,000	6	0.357	0.565	502	1.060	1.475	1.246	3
4,000	8	0.357	0.565	506	1.0685	1.4334	1.1067	4
Pu-U^{238} cases								
2,500	1.5	0.267	0.476	860	1.12	1.459	1.199	5
2,500	1.5	0.244	0.441	850	1.108	1.459	1.263	6
4,000	0	0.404	0.660	427	0.903	1.5947	1.4709	7
4,000	3	0.357	0.565	513	1.055	1.412	1.0764	8
4,000	3	0.404	0.660	540	1.11	1.412	1.0733	9
4,000	3	0.500	0.748	563	1.157	1.411	1.096	10
5,000	3	0.433	0.621	444	1.145	1.443	1.0815	11
6,000	3.5	0.535	0.765	368	1.136	1.463	1.100	12
6,000	4	0.60	0.814	375	1.154	1.444	1.085	13
6,000	5	0.60	0.814	340	1.037	1.406	1.146	14
8,000	0	0.632	0.847	205	0.82	1.604	1.419	15
8,000	6	1.0	1.0	294	1.178	1.434	1.098	16
Pu-U^{238} cases-Pu with 16 % Pu^{240}								
4,000	3	0.41	0.52	575	1.099	1.4162	1.0777	17
4,000	3	0.49	0.66	606	1.16	1.4144	1.0963	18
Pu-Th case-Pu with 6 % Pu^{240}								
3,000	3	1.0	1.0	735	0.9794	1.4860	1.5314	19

BOL = Beginning of Life
EOL = End of Life

In this analysis no reprocessing has been considered, because the amount of fissile material is very low at end of life. The Pu^{240} gets converted almost completely into Pu^{241} and burnt. The other fertile material (Th or U^{238}) is never present in large quantities, and its purpose is more to keep the reactivity at a reasonable level and reduce the changes of power shape during life, than to contribute to fifa by increasing the conversion.

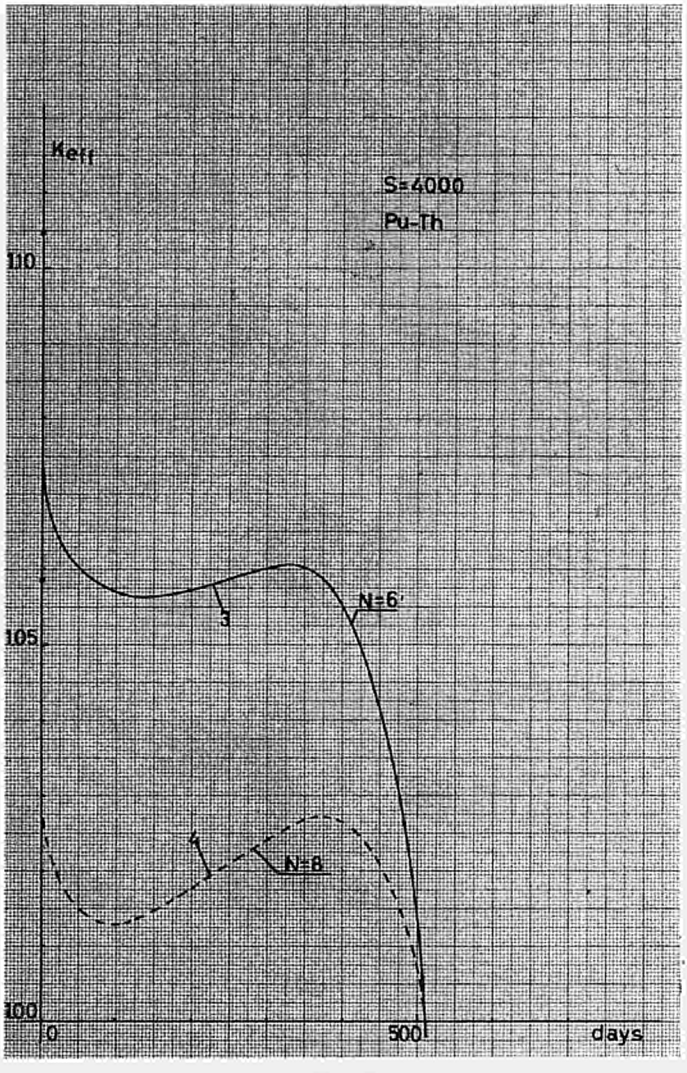

FIG. 9.

As the Pu^{239} has a much higher fission cross section than U^{233}, U^{238} may have some advantage over Th as additional fertile material, when no reprocessing is considered. The conversion ratio is not very different in the two cases, and it is obvious that a higher fission cross section will lead to a longer life.

In the cases considered here the difference in fifa between U^{238} and Th is not great (see table 2 and Fig. 18).

2.1. — POWER DISTRIBUTION.

In cases in which no lumping in the fuel has been considered (homogeneous cell) and the S value is rather low (cases 1 and 2 of table 2) the fuel is burnt as a

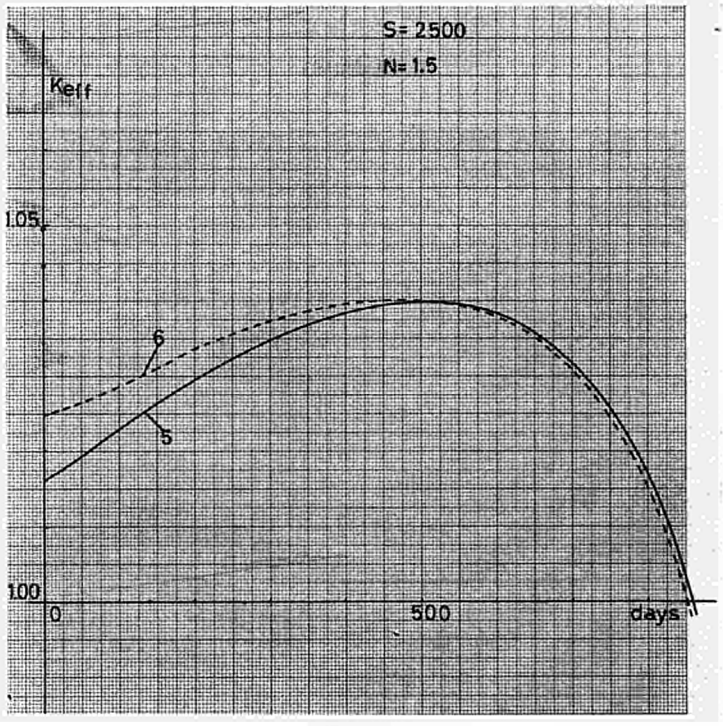
S= 2500
N= 1.5
Keff
1.05
1.00
6
5
0
500
days

FIG. 10.

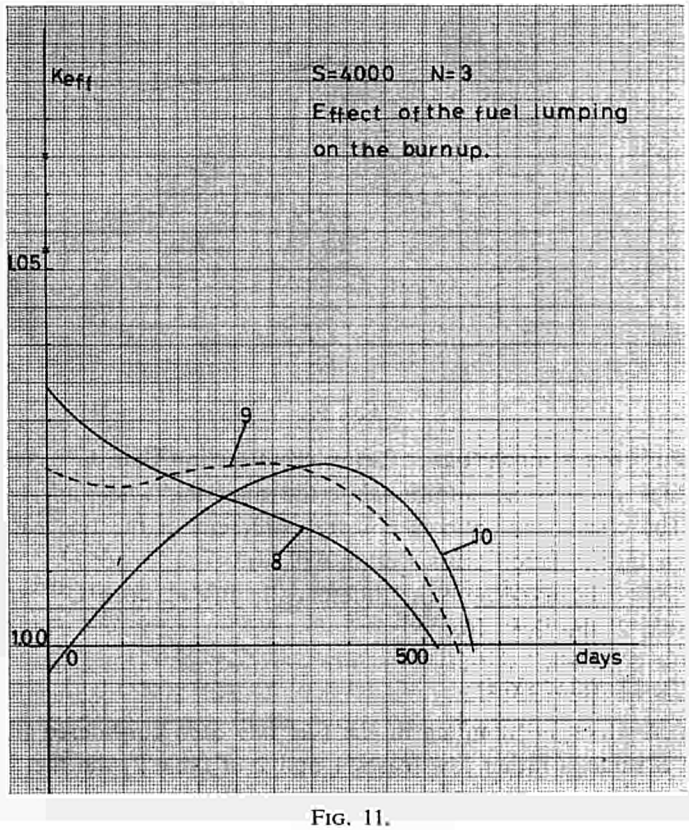
Keff
S=4000 N=3
Effect of the fuel lumping
on the burnup.
1.05
1.00
9
8
10
0
500
days

FIG. 11.

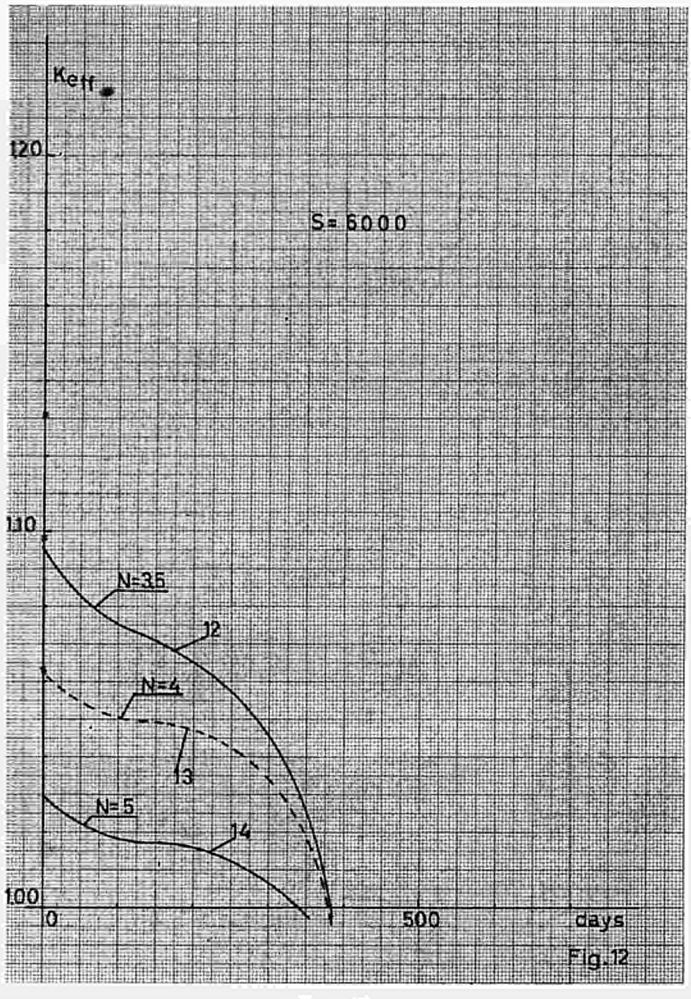

FIG. 12.

rather high speed, but the conversion of Pu^{240} is also very high and the total amount of fissionable material does not change too rapidly until all the Pu^{240} has disappeared.

At that moment the power released by the fuel element drops suddenly. Toward the end of life those elements which were in zones of higher flux reach this point first, and the power distribution in the reactor gets strongly distorted (see the high power shape factors at end of life in table 2, cases 1 and 2, and in Fig. 16).

This fact is avoided by lumping the fuel and adding other fertile material which provides some additional fuel in all the fuel elements.

It is indeed possible to obtain very good power distributions which improve steadily during life, by proper use of lumping and of additional fertile material (see Fig. 17).

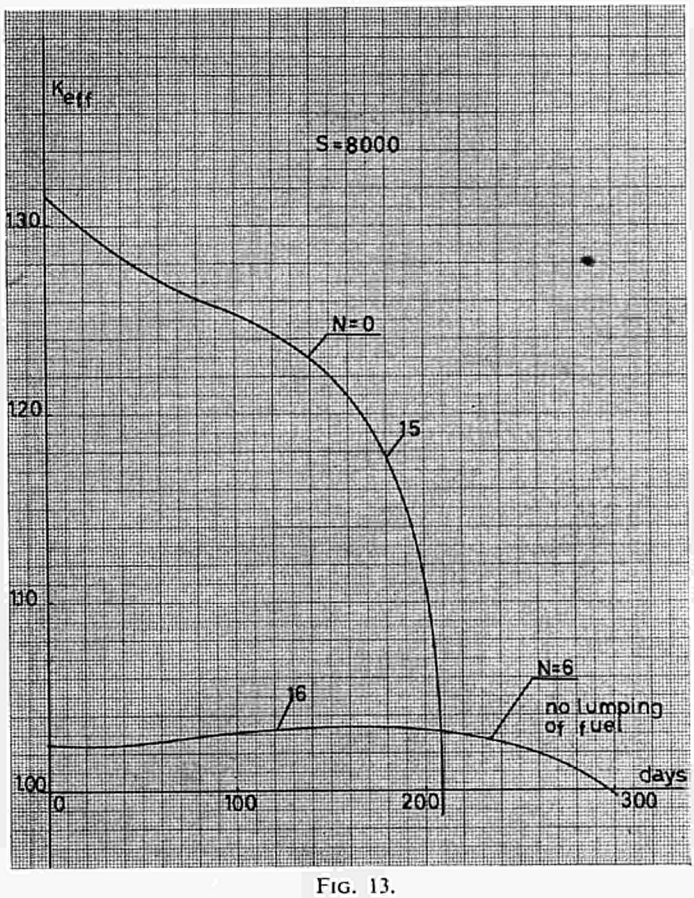
K_{eff}
S=8000
1.30
1.20
1.10
1.00
N=0
15
16
N=6
no lumping of fuel
0
100
200
300
days

FIG. 13.

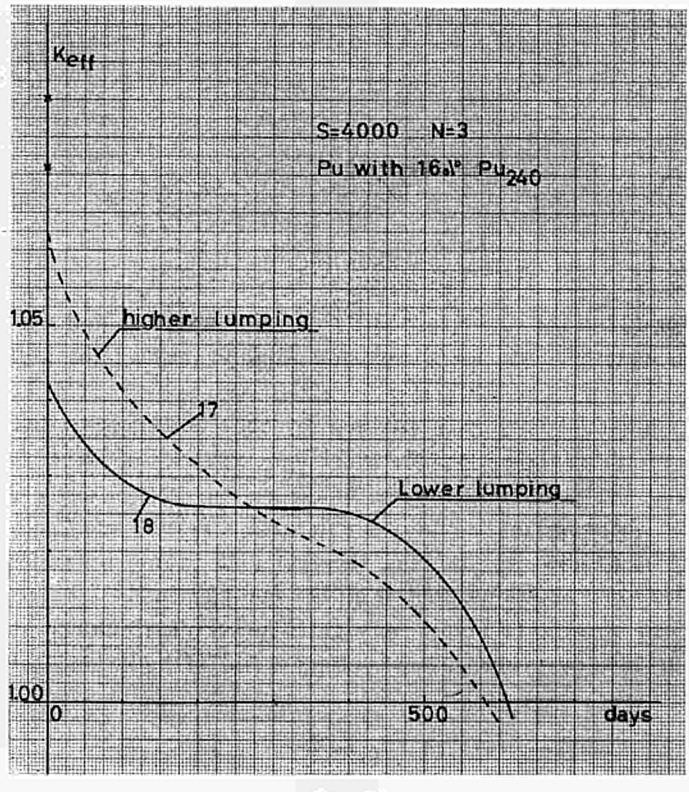
K_{eff}
S=4000 N=3
Pu with 16% Pu_{240}
1.05
1.00
higher lumping
17
18
Lower lumping
0
500
days

FIG. 14.

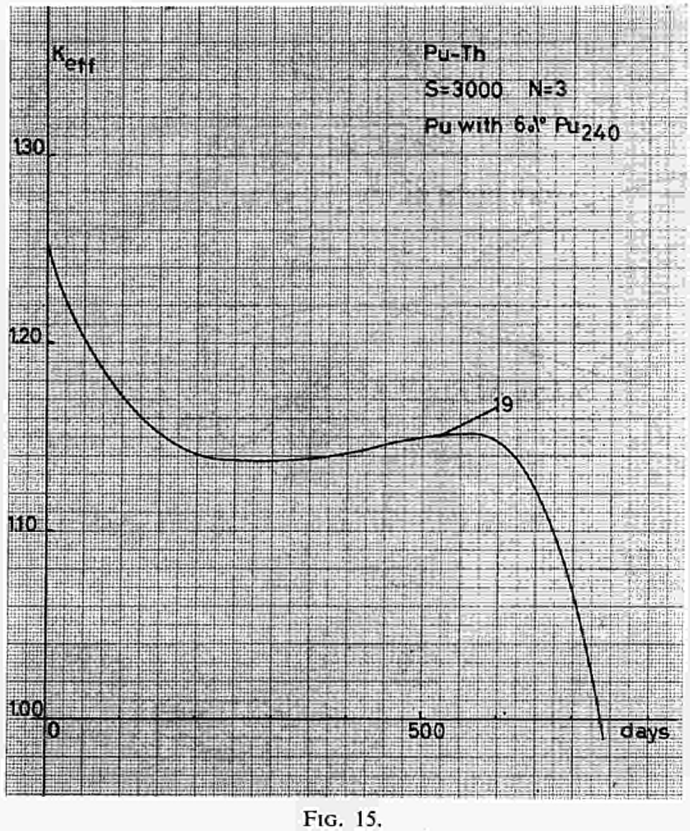
K_{eff}
Pu-Th
S=3000 N=3
Pu with 6.1° Pu_{240}
130
120
110
100
19
0
500
days

FIG. 15.

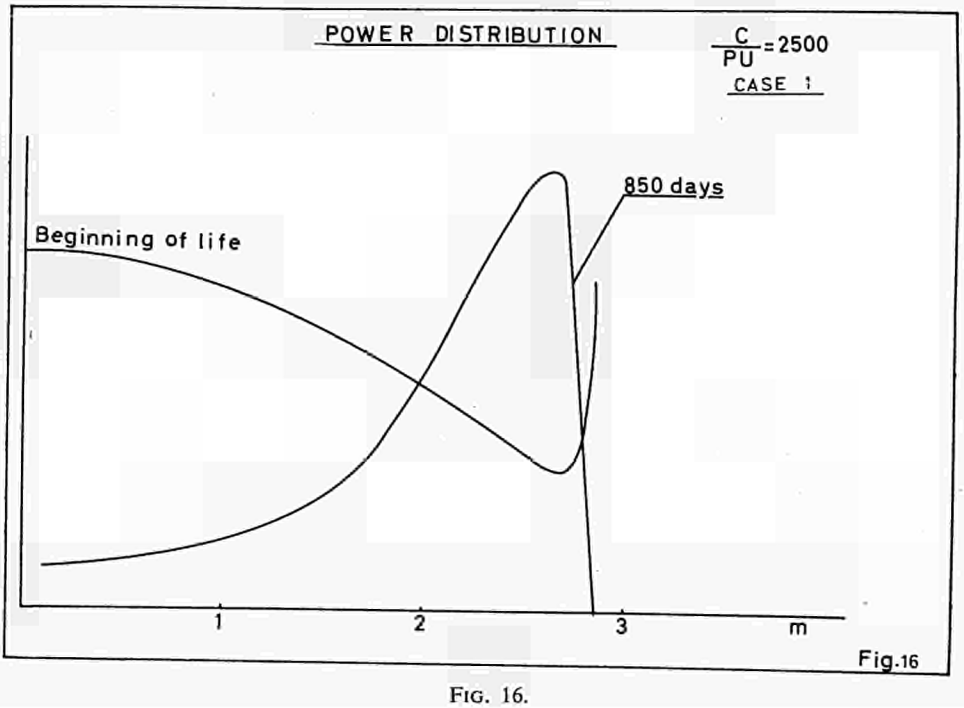
POWER DISTRIBUTION
C/PU = 2500
CASE 1
850 days
Beginning of life
1
2
3
m
Fig.16

FIG. 16.

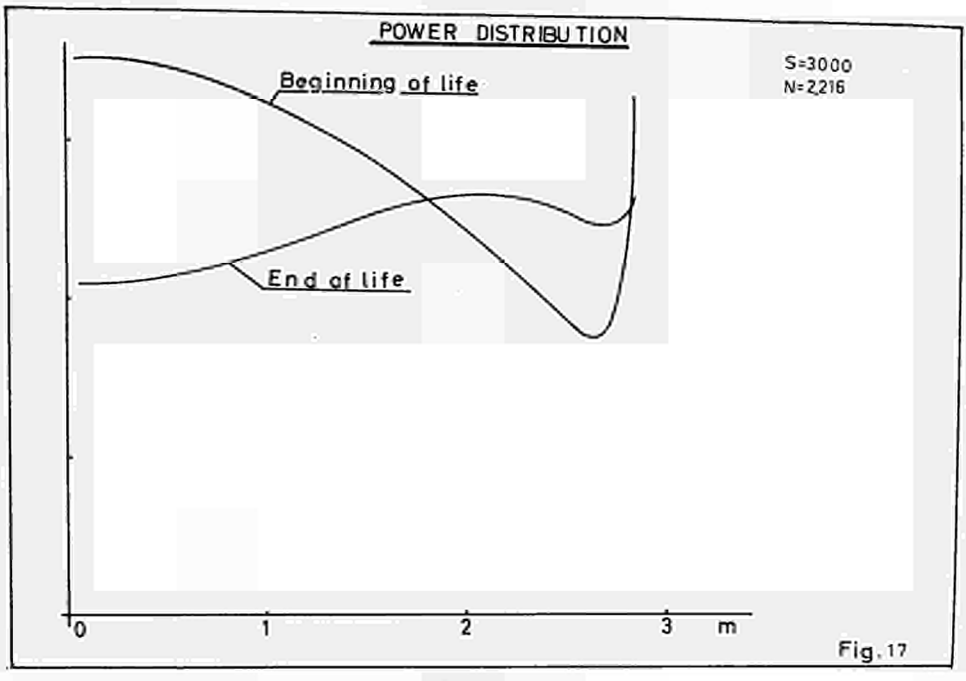

FIG. 17.

2.2. — TYPE OF PU CONSIDERED.

Most of the calculations have been performed with Pu with the following composition

$$Pu^{239} = 65.7\ \% — Pu^{240} = 18.8\ \% — Pu^{241} = 13\ \% — Pu^{242} = 2.5\ \%$$

A few calculations have also performed with Pu with 16 % and with 6 % of Pu^{240} (cases 17, 18 and 19 of table 2). Curves are shown in Fig. 9 to 15.

Plutonium with lower Pu^{240} content requires lower lumping for obtaining flat burn-up curves.

2.3. — TEMPERATURE COEFFICIENT.

For case 9 of table 2 (S = 4,000, N = 3) the temperature coefficient has been calculated for the fuel and for the moderator at beginning and at end of life, at operating temperature. The results were the following :

Beginning of life unpoisoned

$$\text{total } \frac{1}{k}\frac{dk}{dt} = -9.343 \times 10^{-5}$$

$$\text{fuel } \frac{1}{k}\frac{dk}{dt} = -2.869 \times 10^{-5}$$

Beginning of life with Xe and Sm

$$\text{total}\ \frac{1}{k}\frac{dk}{dt} = -8.7868 \times 10^{-5}$$

$$\text{fuel}\ \frac{1}{k}\frac{dk}{dt} = -2.814 \times 10^{-5}$$

End of life (500 days)

$$\text{total}\ \frac{1}{k}\frac{dk}{dt} = -9.312 \times 10^{-5}$$

$$\text{moderator}\ \frac{1}{k}\frac{dk}{dt} = -7.398 \times 10^{-5}$$

It appears that the strong absorption of Pu^{240} is sufficient to keep the temperature coefficient well negative at every moment of the reactor life.

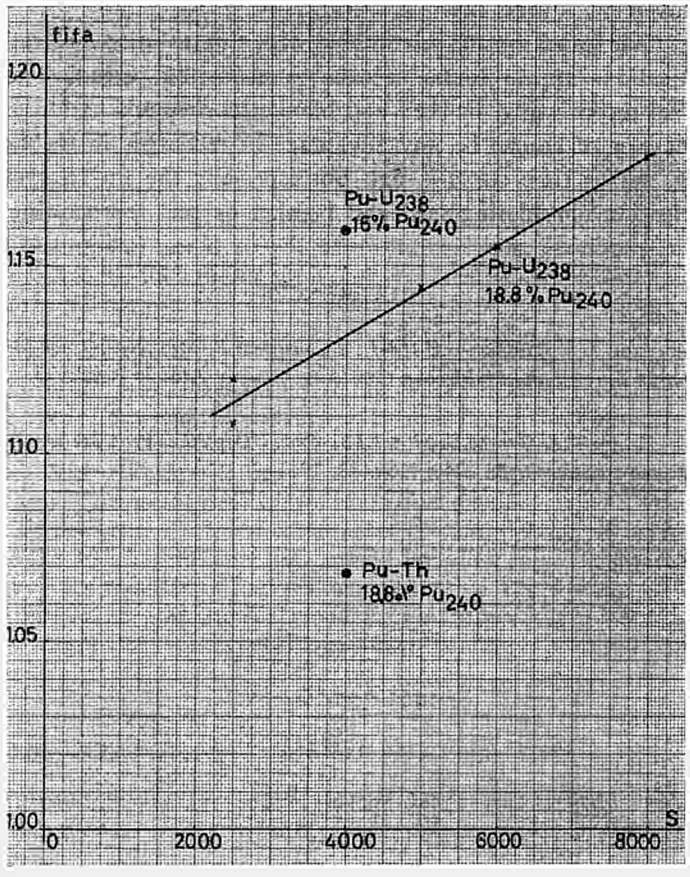

FIG. 18.

2.4. — Conclusion on the Batch Loading Studies.

As it can be seen from table 2 there is no great variation in the number of fissions per initial fissile atom obtainable from the different cases. The fifa value tends to increase for higher S values.

The results are summarised in Fig. 18 where is plotted the best fifa obtainable from each S value. The curve appears to be almost a straight line, but in many cases the differences in fifa can be within the uncertainties of the calculations.

The best solution can be found according to the fuel element design which will be choosen, and the self-shielding obtainable with it.

No S values higher than 8,000 have been considered because beyond that it would not be possible to obtain flat reactivity versus time curves, even with a completely homogeneous reactor and no fuel lumping, except by using Pu with a Pu^{240} content higher than the maximum considered in these calculations.

It appears that one can build rather simple reactors, with flat reactivity versus time curves, and good power distribution, with batch loading without the need of any fuel movement during life.

On the other hand the fifa obtainable with this solution are rather low, and more investigations appear to be necessary considering periodical or continuous recharges, fuel reprocessing, or seed and blanket concepts.

There cannot of course be any advantage in recharges in the cases in which the reactivity versus time curve is flat, but especially the Pu-Th cycles may need more investigation considering seed and blanket or fuel reprocessing.

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THE USE OF LOW-ENRICHMENT URANIUM AND PLUTONIUM IN THE HTGR (1)

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ABSTRACT

The fuel cycle characteristics and fuel cycle costs for a 1,000 MW(e) High Temperature Gas-Cooled Reactor (HTGR) have been extensively studied using U^{233} recycle with U^{235} make-up. In this study, two alternative fuel cycles were investigated :
1) The use of slightly enriched uranium with no recycle.
2) The use of U^{233} recycle with reactor grade plutonium make-up.

The basic configuration of the reactor core, control system, fuel element design and heat removal system were modified as little as possible to accommodate these alternative fuels.

There are two principal reasons for considering alternative fuel cycles :
1. The reactor operator is afforded flexibility in his sources of supply;
2. The U^{235} in slightly enriched uranium and fissile plutonium is valued at considerably less than the U^{235} in fully enriched uranium.

In general, these alternative fuels can be efficiently utilized in the HTGR but at a fuel cycle cost penalty of roughly 0.1 m/kWh.

The slightly enriched uranium fuel cycle was surveyed with the parameters being fuel element residence time, carbon to U^{238} ratio and pin diameter. For fuel residence times of 2 years and 1 inch diameter fuel rods, enrichments below 2 % are feasible and result in burnups of about 10,000 MWD/t. The more economic fuel cycles use fuel pins from 0.2 to 0.3 inch diameter, 3 to 4 year fuel residence times, achieve about 15,000 MWD/t and require fuel enrichments of 3 to 4 %. The effect of linear fuel rating was also considered; at ratings below 2.5 kW/ft, the fuel cycle cost increases very rapidly due to the choice of 0.2 inch as a practical lower limit on fuel pin diameter.

The potential for using Pu make-up in the HTGR is largest when relatively small quantities are used; i.e. when the conversion ratio is high. The Pu make-up fuel cycle was surveyed with the parameters being type of fuel element, fuel loading, and fuel residence time. With all graphite fuel elements roughly 200 kg of Pu are required to operate for a year. The use of BeO spines in the fuel element reduces this requirement to about 100 kg per year. Conversion ratios of 0.8 and 0.9 are achieved in practical fuel cycles for the all graphite and BeO spined fuel elements, respectively. The most economic fuel cycles occur with fuel residence times of 4 to 5 years with the lightest fuel loadings. For an all graphite fuel element, fuel cycle costs of 0.9 m/kWh appear attainable; the use of BeO spines results in a cost increase of about 0.1 m/kWh.

1. — INTRODUCTION

1.1. — GENERAL DESCRIPTION OF THE HTGR AND FUEL CYCLE.

The high-temperature gas-cooled reactor (HTGR) concept being developed at General Atomic is an all-ceramic, thermal-neutron-spectrum reactor using

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helium as the coolant and graphite or a combination of graphite and beryllium oxide as the moderator. For the thorium fuel cycle, the fission products are retained entirely within the pyrolytic-carbon-coated fuel particles. Comparatively little design work has been done on a fuel element suitable for a low-enrichment fuel cycle; the fission products would be retained either by the fuel matrix itself or by purging through the graphite fuel body or by a combination of these. The nominal coolant conditions are :

Coolant inlet temperature	380° C
Coolant outlet temperature . . .	800° C
Coolant pressure	30 atm
Net thermal efficiency	45 %

The core, primary system, and the steam generators are contained within a prestressed concrete reactor structure. A detailed description of the 1,000-MW(e) HTGR [1] can be found elsewhere.

The reactor operates on a graded fuel cycle with semi-annual refueling and a scatter loading. Some consideration is being given to fuel shuffling, but these results are not yet complete. In this report we deal principally with the fuel cycle characteristics and the fuel cycle costs of a 1,000-MW(e) HTGR fueled with either low-enrichment uranium, in the form of oxide or carbide, or thorium with recycled uranium and plutonium make-up. This latter case is a nominal deviation from the standard HTGR fuel cycle, which uses thorium with recycled uranium and U^{235} makeup. Therefore, we have utilized the standard, loose-bed, coated-particle fuel element, which is a graphite cylinder 6.1 m in length (4.7 m active length) and 11.8 cm in diameter. The fuel particles are contained in 14 holes roughly 1.3 cm in diameter within the graphite element. When BeO spines are considered, they are placed in 6.8 cm diameter holes in the center of the graphite fuel element. The fuel elements are arranged in the core in a hexagonal array with a 11.9 cm pitch. All the heat is transferred from the fuel particles to the graphite element and thence to the coolant. No detailed design work has been done on a fuel element suitable for the low-enrichment cycle. For the purpose of establishing the potential of this fuel cycle, it was assumed that stacks of ceramic fuel pellets replace the coated particles in the fuel holes.

The economic parameters used in computing the fuel cycle costs in this study are given in Table 1.1. While the authors feel the tabulated costs are generally consistent with values being projected in the U. S., we recognize the degree of uncertainty in the various values, and we will indicate in the following sections the sensitivity of the cost components to these parameters. In general, those parameters having the greatest uncertainty at this time reflect the least sensitivity to the fuel cycle cost in the HTGR.

1.2. — Interest in Alternative Fuels.

The two principal reasons for considering the alternative fuels (low enrichment and Pu) are (1) the added flexibility afforded the reactor operator and (2)

TABLE 1.1 — Standard Economic Parameters

	Low Enrichment	Makeup Plutonium
Cost per graphite log, $	200	200
Cost of fuel, $/kg Th + U + Pu	26-50[a]	100
Cost of BeO, $/kg	—	26
Cost of recycling BeO, $/kg	—	9
Fabrication losses, %	1	1
Cost of shipping fuel, $/kg Th + U + Pu	10	10
Cost of fuel reprocessing plant, $/day	23,500	23,500
Maximum reprocessing rate, kg/day	1,000	~500
Minimum turnaround time, days	2	2
Maximum turnaround time, days	8	8
Reprocessing losses, %	1	1
Uranium ore cost, $/lb U_3O_8	5	5
Separative work cost, $/kg U	30	30
UF_6 conversion, $/kg U	2.75	2.75
U^{233} value/U^{235} value ($\varepsilon = 0.90$)	—	14/12
Fissile Pu value/U^{235} value ($\varepsilon = 0.90$)	10/12	10/12
Working capital charge, %/year	10	10

[a] The cost of oxide or carbide fuel bodies was assumed to be $25/kg for preparation and $25/kg for pelletizing and assembly with a 0.2-in. — diameter fuel body. This latter charge was taken proportional to the quantity of pellets required. It should be emphasized that these figures are not based on an actual design.

the fact that U^{235} in low-enrichment fuel and plutonium are valued at considerably less than U^{235} when fully enriched. In Fig. 1.1 we show the calculated value of U^{235} as a function of enrichment for several uranium ore costs. We feel that $5/lb of ore should be available in the near future, but as the nuclear capacity grows, the price of ore could quickly increase to $8-10/lb. On the basis of the prediction [2] that uranium could be recovered from sea water for less than $20/lb, we have considered this value an upper limit. It is quite clear that for all ore costs the cost of fissionable material increases very rapidly with enrichment to about 5 %, and from that point on only at a relatively slow rate. Thus, there could be large savings in fuel cycle costs if the very inexpensive U^{235} in low-enrichment fuel could be used to produce energy as well as plutonium.

Two components of the fuel cycle cost are dependent on the value of the U^{235}; the fuel inventory cost and the depletion cost. In Fig. 1.2, we show the envelope of minimum values of the sum of fuel inventory and depletion as a function of initial enrichment for all the fuel cycles surveyed in this study. These points generally represent quite low or minimum fuel cycle costs as well. The same quantities for

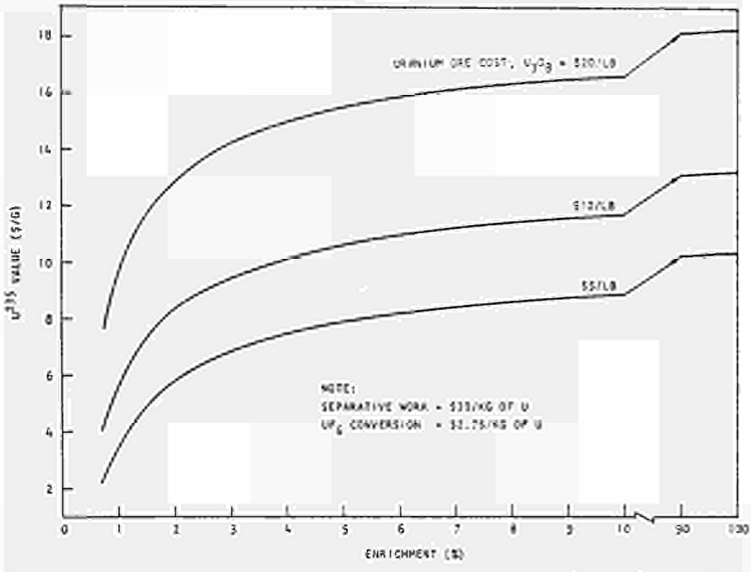

FIG. 1.1. — U^{235} value as a Function of Enrichment for Various U_3O_8 Costs.

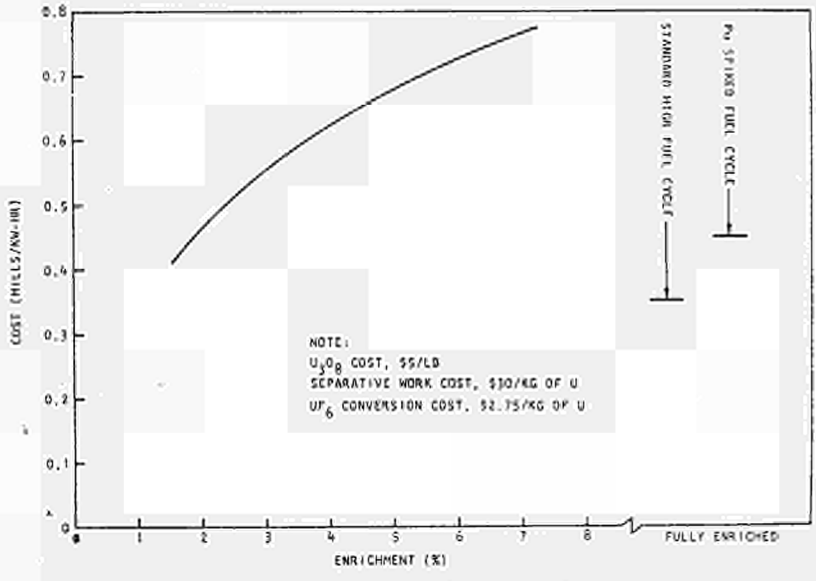

FIG. 1.2. — Sum of Fuel Inventory and Depletion Costs as a Function of Enrichment and Fuel Cycle.

the standard HTGR fuel cycle and the plutonium makeup fuel cycle are shown as well. We see that the standard HTGR fuel cycle using U^{235} makeup has the best potential for low fuel cycle costs. The low enrichment and Pu makeup cycle exhibit costs about 0.1 mill/kW-hr larger; it should be noted that the lowest-enrichment

cases lead to high total fuel cycle cost because of the short residence time and generally require very high heat ratings, which will probably make them difficult to utilize. This is covered in greater detail in the following section. As the cost of uranium ore increases, the sum of fuel inventory and depletion rises most rapidly in the low-enrichment fuel cycle and least rapidly in the standard HTGR fuel cycle. Thus, the 0.1 mill/kW-hr advantage of the standard HTGR fuel cycle will continue to increase with time.

1.3. — Uranium Requirements.

A study of the projected nuclear capacity has recently been issued by the Euratom Commission [3]. Based on this projection and an extrapolation from the year 2,000 to 2,020, we have computed what the uranium requirements would be for plutonium-fueled HTGR's and fast breeder reactor; the results are shown in Fig. 1.3. Two HTGR complexes were considered; one utilized the plutonium discharged from a low-enrichment-fueled HTGR using 3 % enrichment in a 0.3-in. fuel body,

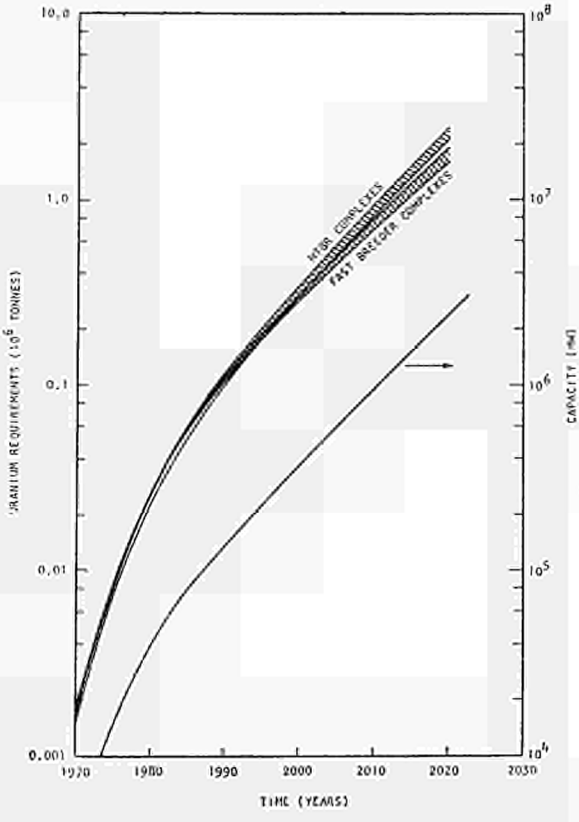

Fig. 1.3. — Installed Capacity and Uranium Requirements for advanced Converter and fast Breeder Complexes.

or pin, with a 3-year residence time. The second system utilized the plutonium discharged from an Organic Cooled Heavy Water Reactor [4] (OHWR). In both cases the plutonium was fed to a BeO-spined HTGR with a Be/Th ratio of 44 and a 4-year residence time. The plutonium for the fast breeder reactor complexes come from the OHWR. Two fast breeder reactors were considered that differed only in their breeding ratios; i.e., 1.3 and 1.4 were used. Both had an initial specific power of 700 kW/kg and a 3.5-year fuel residence time. It is quite interesting that the difference in the uranium requirements among the four reactor complexes is small and within the uncertainties of the data. Thus, any of these complexes would be suitable from a uranium conservation view and the choice would be made on economic considerations alone.

This picture could change materially if a fast breeder reactor with much shorter doubling times were developed. At General Atomic, we are studying a gas-cooled fast breeder that could ultimately obtain a specific power of 1,500 kW/kg and a breeding ratio of 1.5; however, it could be several years before these conditions are achieved.

2. — Low-Enrichment Uranium Cycle

2.1. — Thermal Considerations.

The fuel cycle analysis of HTGR systems using low-enrichment uranium cannot be completely dissociated from heat-transfer considerations, since a close interdependence exists between nuclear, thermal, and economic parameters. In this study it was assumed that the heat is generated in stacks of ceramic fuel pellets and then transferred from the stacks to the surrounding graphite and from the graphite to the coolant gas. For typical fuel element designs, the average graphite temperature is mainly a function of the core power density (W/cm^3), while the temperature drop inside the fuel pellets is primarily determined by the thermal conductivity of the fuel and by the linear rating (kW/ft).

The temperature difference between the centerline and the surface of a fuel pellet is, to a first approximation, independent of the pellet diameter. From the thermal design viewpoint, it is therefore desirable to divide the fuel into as many fuel pellet stacks as possible in order to maximize the total length of fuel stacks and to minimize the linear rating. However, the nuclear and economic characteristics tend to be more favorable for fewer stacks of large diameters, since they lead to lower enrichments as a consequence of the stronger self-shielding in the U^{238}.

Because of the low thermal conductivity of uranium oxide, its use as fuel in low-enrichment HTGR's can necessitate quite severe design restrictions. For a power density of 7 W/cm^3 and a maximum fuel temperature limit of 1,500° C, the maximum linear rating, i.e. the average rating times the radial peaking factor (1.2) times the maximum age peaking factor, which is calculated for each case and is defined as the ratio of the maximum-to-average power generation rate of an element, should be below 2.5kW/ft. This low linear rating cannot be achieved without

TABLE 2.1. — Fuel cycle characteristics for low-enrichment HTGR's

Fuel Cycle Length (years)	C/U^{238}	Initial Enrichment (%)				Final Enrichment (%)				Fissile Pu Discharged (kg)			
		Pin Diam (in.)				Pin Diam (in.)				Pin Diam (in.)			
		0.2	0.3	0.5	1.0	0.2	0.3	0.5	1.0	0.2	0.3	0.5	1.0
2	125	3.65	2.62	2.02	1.69	2.29	1.36	0.83	0.54	137.5	105.2	79.2	60.5
	175	2.65	2.29	2.09	1.99	1.02	0.69	0.48	0.36	71.8	55.7	43.8	35.4
	225	2.68	2.55	2.50	2.49	0.62	0.46	0.35	0.28	43.7	35.4	29.1	24.5
	275	3.05	3.03	3.04	3.08	0.46	0.36	0.29	0.24	30.4	25.4	21.6	18.6
3	125	4.64	3.32	2.55	2.17	2.64	1.48	0.83	0.49	123.8	90.4	63.9	46.0
	175	3.41	2.98	2.77	2.71	1.03	0.63	0.40	0.27	58.6	43.1	32.5	25.5
	225	3.57	3.47	3.46	3.50	0.54	0.37	0.26	0.19	33.2	26.0	20.9	17.2
	275	4.19	4.22	4.30	4.40	0.35	0.25	0.18	0.14	22.3	18.3	15.2	12.9
4	125	5.67	4.05	3.13	2.70	3.01	1.62	0.84	0.46	115.5	81.6	54.9	38.7
	175	4.22	3.73	3.53	3.50	1.07	0.61	0.36	0.22	50.9	36.0	26.4	20.2
	225	4.54	4.45	4.49	4.59	0.50	0.31	0.20	0.13	27.4	21.0	16.6	13.5
	275	5.41	5.50	5.64	5.80	0.28	0.18	0.13	0.09	18.0	14.5	12.0	10.1
5	125	6.72	4.83	3.74	3.27	3.39	1.78	0.87	0.44	109.7	75.8	49.0	32.5
	175	5.09	4.54	4.33	4.33	1.14	0.60	0.32	0.18	46.0	31.5	22.5	16.9
	225	5.54	5.49	5.58	5.73	0.47	0.27	0.16	0.10	23.8	18.0	14.0	11.2
	275	6.69	6.83	7.03	7.24	0.24	0.14	0.09	0.06	15.4	12.3	10.0	8.3

reducing the specific power below its optimum, since there are practical limits set to the subdivision of the fuel. For the present study, a pellet diameter of 0.2 in. was considered a lower limit. The thermal design problems are greatly eased if carbide fuel is used. Owing to its higher thermal conductivity, considerably higher linear ratings (in the range of 10 kW/ft) are acceptable.

2.2. — Fuel Cycle Characteristics.

The main difference between the thorium and the low-enrichment cycle arises from dependence of the fuel value on the fissile-to-fertile ratio in the latter cycle. Because of this dependence, a fuel element concept based on coated particles is not suitable for a low-enrichment system. The carbon coating introduces a significant amount of moderating material in the packed bed, which subsequently requires enrichments in excess of 10 %. Thus the value of the U^{235} is nearly as large as in fully enriched uranium, which together with the inherently lower conversion ratio of thermal uranium-plutonium systems, makes the concept economically unattractive.

A higher degree of lumping can be achieved by using oxide or carbide fuel bodies. For such a fuel concept, enrichments of 4 % and less can be used, thereby reducing the cost of the U^{235} considerably. As mentioned in the previous section, there are limits set on the degree of lumping that can be achieved in an HTGR system.

The fuel cycle characteristics of a low-enrichment HTGR system have been determined for a range of fuel loadings (C/U^{238} ratios), fuel rod diameters, and fuel residence times. The calculations were based on UO_2 fuel; however, as far as nuclear aspects are concerned, essentially the same conclusions apply for carbide fuel. Some fuel cycle parameters of the systems studied are listed in Tables 2.1 and 2.2. Typical values for the initial enrichment are between 2 % and 4 % and between 0.5 and 0.65 for the conversion ratio. Long fuel residence times require rather high enrichments, and it is expected that the optimum cycle will be shorter than for a thorium-fueled HTGR using uranium recycle with U^{235} makeup.

The lower limit for the initial enrichment is primarily set by the power density. Present HTGR designs call for power densities of the order of 7 W/cm^3. It is clear that, by reducing this value, savings in burnup charges can be achieved. However, it was concluded that, when balanced against the higher capital cost of the reactor necessitated by the larger core, a net increase in generating cost would result. Another factor affecting the enrichment requirements of an HTGR is the inherently high moderator temperature. For the purpose of calculating the nuclear group cross sections, the average fuel temperature was assumed to be 1,000° C and the average graphite temperature 900° C. A reduction of 300° C in both temperatures led to only marginal changes in the fuel cycle and economic characteristics.

2.3. — Fuel Cycle Cost.

In the HTGR system fueled with uranium of low enrichment, as pointed out in the previous section, there exists a much stronger relationship between nuclear,

TABLE 2.2. — Fuel cycle characteristics for low-enrichment HTGR's

Fuel Cycle Length (years)	C/U^{238}	Conversion Ratio				Burnup (MWd/Ton)				Average Specific Power			
		Pin Diam (in.)				Pin Diam (in.)				Pin Diam (in.)			
		0.2	0.3	0.5	1.0	0.2	0.3	0.5	1.0	0.2	0.3	0.5	1.0
2	125	0.669	0.665	0.652	0.633	7,200	7,300	7,300	7,300	810	1,210	1,710	2,240
	175	0.640	0.621	0.599	0.571	10,200	10,200	10,200	10,300	1,860	2,380	2,900	3,370
	225	0.596	0.570	0.543	0.515	13,100	13,100	13,100	13,100	2,890	3,360	3,780	4,140
	275	0.549	0.522	0.495	0.467	15,900	15,900	15,900	15,900	3,690	4,070	4,400	4,680
3	125	0.650	0.648	0.636	0.615	10,700	10,800	10,900	11,000	650	1,000	1,460	1,970
	175	0.627	0.606	0.580	0.551	15,200	15,200	15,300	15,300	1,570	2,060	2,550	2,990
	225	0.581	0.554	0.524	0.495	19,500	19,500	19,500	19,500	2,500	2,940	3,340	3,680
	275	0.533	0.505	0.476	0.448	23,600	23,600	23,600	23,600	3,220	3,590	3,900	4,180
4	125	0.629	0.631	0.619	0.596	14,100	14,300	14,500	14,500	540	850	1,270	1,750
	175	0.611	0.589	0.562	0.531	20,000	20,200	20,200	20,200	1,350	1,800	2,260	2,670
	225	0.564	0.537	0.506	0.475	25,700	25,700	25,700	25,700	2,180	2,600	2,980	3,310
	275	0.517	0.488	0.458	0.428	31,100	31,100	31,000	31,000	2,850	3,200	3,500	3,750
5	125	0.609	0.614	0.602	0.578	17,400	17,800	18,000	18,100	460	730	1,120	1,560
	175	0.595	0.573	0.545	0.513	24,800	25,000	25,000	25,000	1,170	1,580	2,020	2,410
	225	0.550	0.521	0.489	0.458	31,800	31,800	31,800	31,700	1,930	2,330	2,690	3,000
	275	0.502	0.472	0.441	0.411	38,400	38,300	38,200	38,100	2,550	2,870	3,160	3,400

economic, and thermal parameters than exists in the standard thorium fuel cycle. Therefore, it is not meaningful to quote cost figures without referring to the thermal aspects of a particular design. For example, large fuel lump sizes lead to the lowest costs; however, they require some type of internal cooling to achieve their full potential.

The minimum fuel cycle costs for the parameter range described previously are shown in Fig. 2.1 as a function of the maximum linear rating. The fuel cycle costs for low-enrichment systems are quite sensitive to the ore cost, because a large fraction of the total cost is associated with the U^{235} value, which is particularly sensitive to ore costs at low enrichment. At low linear ratings, the fuel cycle cost increases sharply in part because of the increased enrichments required for the small lump sizes. A second important factor is the lower practical limit set on the fuel pellet diameter, which was taken as 0.2 in. in this study. Systems with linear ratings below 2.5 kW/ft require specific powers that are much too low to yield attractive fuel cycle costs.

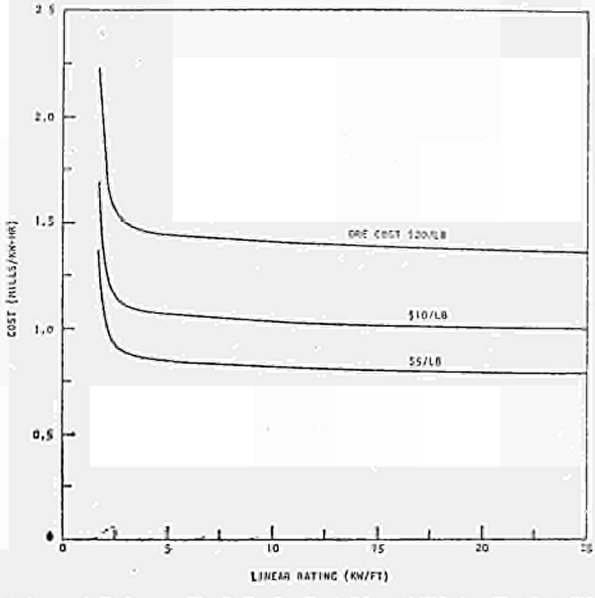

FIG. 2.1. — Minimum Fuel Cycle Cost Versus Linear Rating (kW/ft).

The fuel cycle cost components for two typical cases, one representative of an oxide design (2.5 kW/ft) and one of a carbide design (9 kW/ft), are given in Table 2.3. The sensitivity of these cost components to changes in economic parameters is shown by indicating the percentage change required to change the fuel cycle cost by 0.1 mill/kW-hr. In both cases the fuel cycle cost is fairly sensitive to the ore cost. The case with 2.5 kW/ft is also fairly sensitive to changes in the fabrication cost and the interest rate.

TABLE 2.3. — Sensitivity of Fuel Cycle Cost Components to Economic Parameters

	2.5 kW/ft (0.2 in., C/U^{238} = 175. 2 years)		9 kW/ft (0.3 in., C/U^{238} = 225. 3 years)	
	Cost (mill/kW-hr)	% Increase to Add 0.1 mill/kW-hr	Cost (mill/kW-hr)	% Increase to Add 0.1 mill/kW-hr
Fabrication [a] . . .	0.22	45	0.13	80
Shipping [a]	0.02	475	0.02	610
Reprocessing [a]. . .	0.09	108	0.08	130
Fuel cost [b] :	0.61		0.59	
Ore cost		66		57
Inventory		55		84
Total inventory (including fuel) . .	0.19	53	0.12	82
TOTAL cost . . .	0.95		0.82	

[a] Including working capital charges.
[b] Net depletion plus fuel inventory.

3. — PLUTONIUM SPIKING

3.1. — FUEL CYCLE CHARACTERISTICS.

The HTGR is capable of achieving breeding in large sizes when BeO replaces the graphite in the spine of the fuel element even with complete fission product retention [5]. If the volatile fission products are allowed to escape, a significant breeding gain can be achieved. However, under the standard economic conditions generally used in projecting costs in the U. S. the reduced burnup and specific power required to achieve breeding are not warranted. Therefore, the reactor will most likely be operated in the high-conversion mode which will require fully enriched uranium makeup and segregated fuel [1]. This latter requirement limits the buildup of the higher isotope poisons, U^{236} and Np^{237}, by selectively discharging from the system those portions of the fuel that have experienced the largest burnup. While it does not appear that segregated fuel will add materially to the fuel cycle cost or that the availability of fully enriched uranium will be limited, the use of plutonium discharged from a low converter reactor (nominally taken as 78 % Pu^{239}, 17 % Pu^{240}, 5 % Pu^{241}) for makeup would obviate both requirements. It is presumed that sufficient fully enriched uranium would be available to bring the reactor to equilibrium, at which point all remaining requirements would be made up with plutonium.

The use of discharge plutonium as a fissile material in an HTGR produces some unusual characteristics. The Pu^{239} does not exhibit a particularly large eta

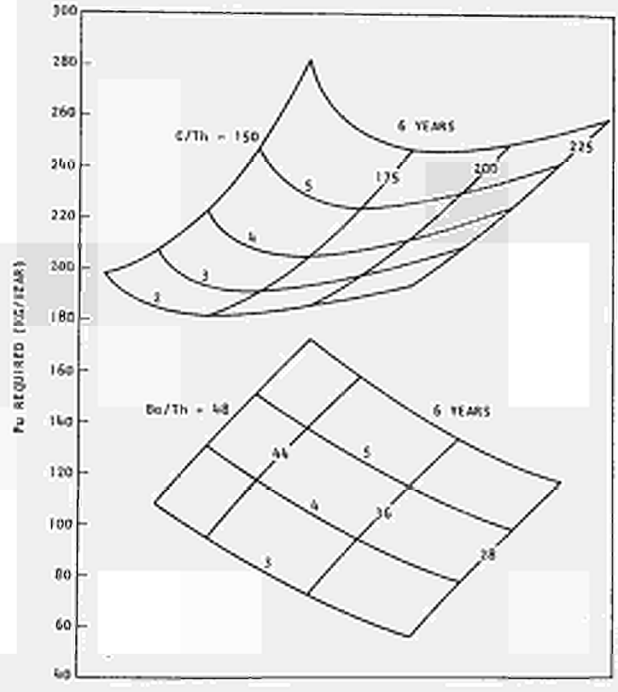

FIG. 3.1. — Annual fissile Pu requirements in 1,000-MW(e) HTGR using Pu makeup.

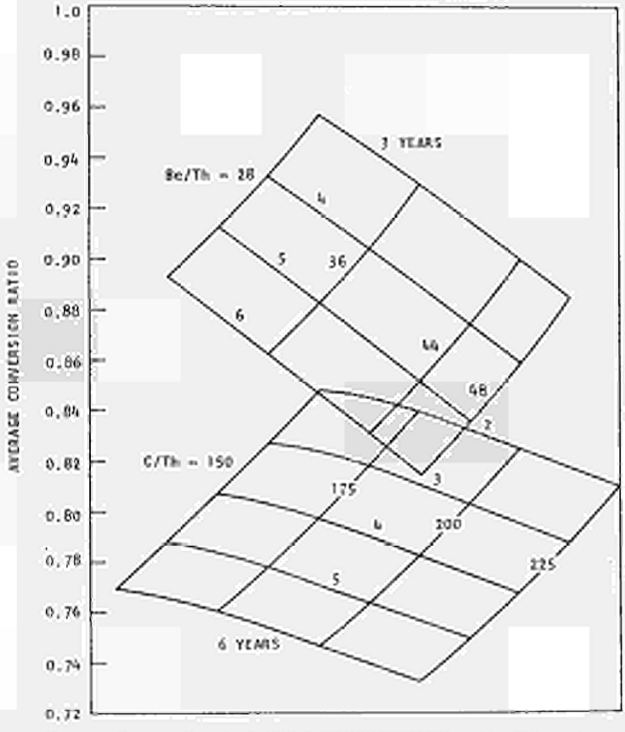

FIG. 3.2. — Conversion ratios in 1,000-MW(e) HTGR using Pu makeup.

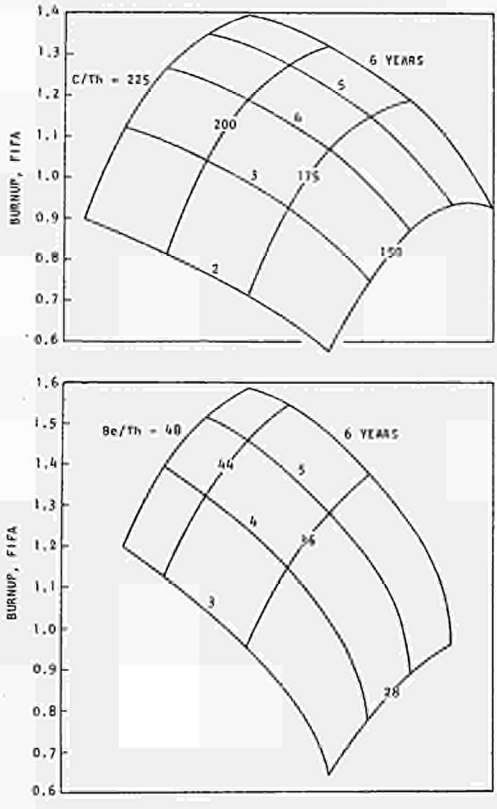

FIG. 3.3. — Fuel burnup in 1,000-MW(e) HTGR using Pu makeup.

in the prevailing spectrum, and the eta has a tendency to decrease rapidly as the spectrum hardens with increasing plutonium concentration. Further, the plutonium carries a strongly absorbing fertile material with it (Pu^{240}), which then leaves very few excess neutrons available for conversion in thorium. Therefore, discharge plutonium can be used to advantage in the HTGR primarily as a makeup fuel with U^{233} producing the majority of the power.

The quantity of plutonium required in kg per year is shown in Fig. 3.1 as a function of moderator-to-thorium ratio, as C/Th or Be/Th, and fuel residence time, in years, for both the all-graphite fuel elements and the BeO-spined fuel elements. Figures 3.2 and 3.3 show the average conversion ratio and the burnup in fissions per initial fissile atom (fifa), respectively, for the same independent variables. We see that the combination of heavy loading and short residence times result in plutonium requirements of less than 100 kg/year and conversion ratios well above 0.9 for the BeO-spined fuel elements. For the all-graphite fuel elements, the highest conversion ratios vary from 0.8 to 0.85, and thus the plutonium requirements increase to roughly 200 kg/year. As will be demonstrated in the following section, the fuel cycle economics dictate light loadings and fuel residence times from 4 to

6 years for all the uranium ore costs considered. Thus, it is most probable that the reactor will be operated with a C/Th ratio of 200 or greater for the all-graphite fuel element and a Be/Th ratio of 36 or greater for the BeO-spined fuel element. The use of the lightest loading may be prohibited by excessively large age peaking factors (1). It will be shown in the following section that a balance between the economics and the heat removal requirements will result in a C/Th ratio of about 200 with a 4-year residence time and Be/Th of about 44 with a 5-year residence time. For these conditions, the conversion ratios are 0.78 and 0.85, the burnups are 1.2 and 1.5 fifa, and the plutonium requirements are 210 and 140 kg/year for the all-graphite and the BeO-spined fuel element, respectively.

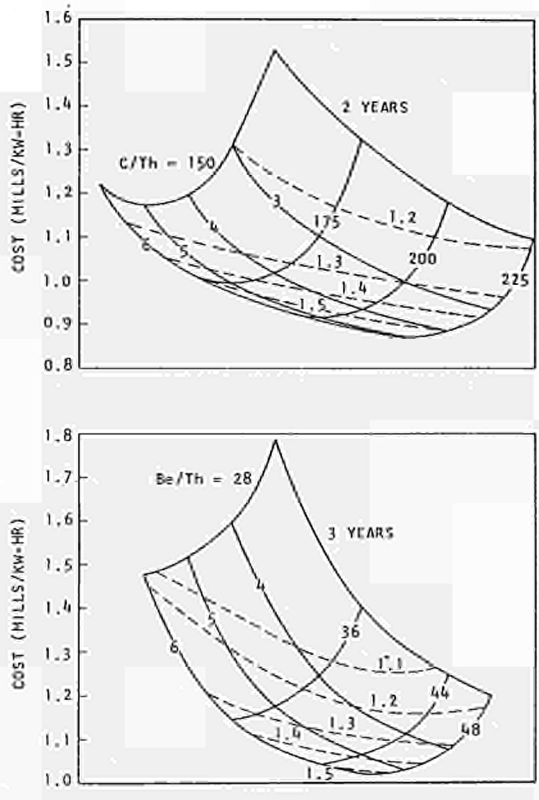

FIG. 3.4. — Fuel cycle costs and age peaking factors in 1,000-MW(e) HTGR using Pu makeup.

(1) The age peaking factor is defined as the ratio of the fission rate in freshly loaded fuel relative to the fission rate in fuel of average exposure in the average reactor flux. This peaking results from using a graded fuel cycle in which fuel of all degrees of exposure are intimately mixed in the core and thus must be added to any general power peaking associated with the gross flux distribution.

TABLE 3.1. — Sensitivity of fuel cycle cost components to economic parameters

	Lowest Fuel Cycle Cost (C/Th = 225. 4 years)		Lowest Pu Requirements (Be/Th = 28. 3 years)		Best Compromise (Be/Th = 48. 5 years)	
	Cost (mills/kW-hr)	% Increase to Add 0.1 mill/kW-hr	Cost (mills/kW-hr)	% Increase to Add 0.1 mill/kW-hr	Cost (mills/kW-hr)	% Increae to Add 0.1 mill/kW-hr
Fabrication [a]	0.17	60	0.57	20	0.24	40
Shipping [a]	0.02	590	0.03	290	0.01	1,000
Reprocessing [a]	0.07	150	0.22	45	0.06	160
Fuel Cost : [b]	0.59		0.71		0.53	
Ore Cost		60		40		60
Interest rate		30		15		30
Total inventory (including fuel)	0.35	30	0.85	10	0.51	20
Total cost	0.89		1.79		1.03	

[a] Including working capital charge.
[b] Net depletion plus fuel inventory.

3.2. — Fuel Cycle Costs.

The equilibrium fuel cycle cost as a function of the variables delineated in the preceeding section are shown in Fig. 3.4 for the case of \$5/lb uranium ore. Superimposed on the figures are the age peaking factors as previously defined. It is generally felt that age peaking factors from 1.3 to 1.4 can be accommodated in the fuel elements being studied in the Target HTGR program [1]. This will result in a minimum fuel cycle cost of about 0.9 mill/kW-hr for all-graphite fuel elements and 1.0 mill/kW-hr for the BeO-spined fuel element. The BeO-spined fuel element consistently produces fuel cycle costs about 0.1 mill/kW-hr larger than the corresponding all-graphite fuel element, primarily because of the large value associated with the BeO, the larger quantity of thorium particles that is required, and a somewhat lower specific power. However, the BeO-spined fuel element is of continued interest because of its excellent fuel conservation characteristics and the potential for achieving breeding in a thermal-spectrum reactor.

The sensitivity of the fuel cycle cost components is indicated in Table 3.1 for three specific cases. It should be noted that the cases yeilding low fuel cycle costs are fairly insensitive to changes in handling costs (fabrication, etc.) and uranium ore costs and are only sensitive to changes in both the fabrication cost and the interest rate. The one economic parameter that is least controllable is the cost of uranium ore and, therefore, this parameter has been pursued further. In Fig. 3.5 we show the minimum fuel cycle cost as a function of uranium ore cost and limiting age peaking factor for both fuel elements. It should be noted that all the cases shown behave similarly and increase by less than 0.5 mill/kW-hr for a factor of four increase in ore cost. We, at General Atomic, generally feel that the projected fuel cycle costs

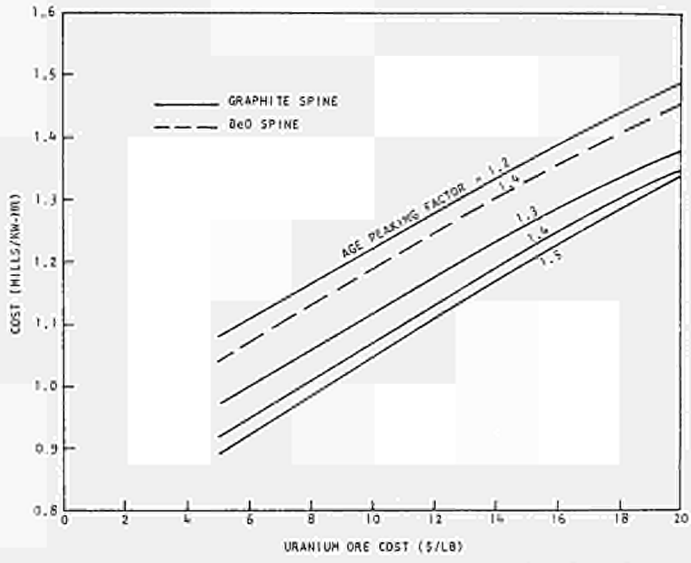

Fig. 3.5 — Minimum fuel cycle cost versus ore cost as a function of age peaking.

must not exceed 1.7 mills/kW-hr for any anticipated ore cost if a reactor concept is to have long-term acceptance. This results from an anticipated cost of coal of \$0.23/10^6 Btu and a \$20/kW advantage in capital cost for the fossil fueled plant. All of the fuel cycle costs shown in Fig. 3.5 meet this critical requirement.

4. — Conclusions

The HTGR concept being developed at General Atomic yields attractive fuel cycle costs on either the low-enrichment uranium fuel cycle or the thorium fuel cycle with plutonium makeup described in this report. The standard thorium fuel cycle using fully enriched uranium makeup exhibits costs from 0.1 to 0.2 mill/kW-hr lower than these fuel cycles. However, the difference could be reduced by a change in the economic parameters or could be accepted because of the availability of low-enrichment uranium or plutonium. Thus, the reactor operator has the flexibility to use any of three fuel cycles depending on the prevailing conditions.

The uranium resources required for the HTGR complexes are modest and quite comparable with reactor complexes based on the fast breeder. For a nuclear capacity expanding with the rapidity of that projected by the Euratom Commission, the resources are primarily dependent on the specific power rather than the conversion ratio. The flexibility of operation available in the HTGR makes it possible to obtain a high specific power during this period of rapid expansion and, subsequently, to operate with a higher conversion ratio as the capacity doubling time stretches out.

APPENDIX

Calculational Model

Fuel cycle calculations were performed using GAFFE [6], a zero-dimensional, multigroup, equilibrium cycle, depletion code that solves the burnup equations analytically assuming a constant flux between reloads. In addition to the Th-U-Pu chains, concentrations of sixteen fission product nuclides are evaluated with the remaining nonsaturating fission products treated as an aggregate.

The spectra, conversion ratio, and other physics parameters are based on the time-averaged nuclide concentrations. To account for the finite reloading interval, the nuclide concentrations at the beginning and end of the six-month refueling intervals are calculated. Based on these concentrations the flux, spectrum, and k_{eff} are re-evaluated at these points. Given the fertile loading and desired fuel residence time, the code iterates on the fissile loading to produce the desired k_{eff} at the end of the reload interval. Excellent agreement is obtained in this manner relative to the more conventional zero-dimensional depletion calculations in which the burnup is based on the spectrum obtained with the nuclide concentrations of the previous time step. The adequacy of the zero-dimensional burnup model for large

HTGR's has also been demonstrated by comparison with one-dimensional calculations.

All of the burnup calculations discussed here were performed using sixteen energy groups (six fast, ten thermal). Fast cross sections were obtained with the aid of the GAM II [7] code, using a Bl approximation for the spectrum calculations and a method developed by L. W. Nordheim for the treatment of resonance absorption. Since the fertile loading and graphite concentration are the prime factors in determining the fast spectrum and are specified as input to the GAFFE code, the fast spectrum used in the burnup calculations is nearly identical to that over which the cross sections were averaged. Minor variations are accounted for by using six energy groups.

One set of microscopic thermal cross sections was used for all of the fuel cycle calculations. The thermal cross sections were averaged over a typical spectrum characterized by $C/U^{235} = 5{,}000$, $C/Pu^{239} = 20{,}000$. This was done with the aid of the GATHER II [8] code using the Bl approximation and the Parks scattering kernel to obtain the spectrum. Use of ten thermal groups is felt to be more than sufficient to provide a satisfactory self-adjusting spectrum. The group structure in the vicinity of the 0.3-ev resonance in Pu^{239} and the 1.05-ev resonance in Pu^{240} was carefully chosen to reflect the energy self-shielding.

The fuel cycle costs were computed by the method outlined by Vallance [9]. While this method does not deal with present worth accounting, it is felt that the results are fairly accurate and will yield correct relative values.

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SESSION IV :

USE OF PLUTONIUM AS INITIAL FISSILE INVESTMENT AND/OR AS MAKE-UP FUEL

Introduction prepared by C. P. GRATTON, Winfrith, U. K. A. E. A.

INTRODUCTION

1. The papers presented to this session of the symposium have many detailed aspects in common and in order to avoid unnecessary repetition, I propose to limit my comments to a general review of the use of plutonium enriched fuels in the HTGC reactor.

2. My material is extracted from the five papers listed in your programme.

(i) Bruneder and Haubert "Plutonium fuel cycle studies for a large high temperature graphite moderated reactor";

(ii) Massimo "Some aspects of the use of plutonium in high temperature reactors";

(iii) Carlsmith "Fuel cycles for high temperature gas-cooled reactors utilizing plutonium";

(iv) Gratton "A review of the studies made on the utilization of plutonium";

(v) Jaye, Fischer, Lee "The use of Low-Enrichment Uranium and Plutonium in the HTGR".

3. A great deal of work has been carried out over the last few years on the fuel cycle performance of High Temperature Reactors based on the Dragon concept. Much of this work has been based on the U^{235}-Th^{232}-U^{233} fuel cycle and we have heard at this symposium the up-to-date situation of these. Calculations have indicated that if a large HTGC programme is to be supported, substantial quantities of highly enriched uranium 235 will be required. With the increased production of plutonium from power reactors, it is most important that this source of fissile material be studied in relation to the High Temperature Reactor.

4. At the outset, I must draw your attention to the fact that unlike the uranium 235 cycle, where everything can be clearly defined, with plutonium it is necessary to define just what one means by plutonium. The civil plutonium extracted from a nuclear power station will contain the higher isotopes and my point is best illustrated in Table I.

This variation depends on the assumed supply of the plutonium and the relative concentrations obtained depend on the fuel irradiation and the spectrum conditions of the source reactor. This variation in isotopic content immediately raises the question as to how sensitive our design proposal is to changes in fuel supply.

TABLE I. — Pu. Isotopic Concentrations Used in Various Studies

	Dragon	Euratom	UKAEA	ORNL	G.A.
Pu^{239}	80	65.7	80	52	78
Pu^{240}	16	18.8	16	29.5	17
Pu^{241}	4	13	4	11.5	5
Pu^{242}	—	2.5	—	7.0	—

REACTOR PHYSICS

(i) *Plutonium Cross-Sections.*

5. In our reactor physics and fuel cycle studies, the performance of the reactor is dominated by the variation of the neutron cross-section in the thermal energy region. The plutonium 239 resonance at 0.3 ev. influences the temperature coefficient of reactivity; as the moderator temperature increases, the absorption in Pu^{239} relative to other isotopes goes up and thereby provides a positive component to the temperature coefficient. We also note that the value of alpha increases within the resonance yielding a value of eta (the number of neutrons produced divided by the number of neutrons absorbed) for plutonium in typical HTR spectrum conditions between 1.75 to 1.8 as compared to around 2.0 for uranium 235.

6. The plutonium 240 resonance at 1.06 ev. is very large and this undergoes very large self-shielding in typical situations.

7. The result of the lower eta for Pu^{239} and the Pu^{240} captures means that in order to achieve criticality, the level of additional fertile material in the core is much less than for a uranium 235 system.

(ii) *Methods of Calculation.*

8. The methods of calculation in all papers are based on multigroup techniques in one form or another. The object, of course, is to derive a satisfactory representation of the neutron spectrum and to follow the changes in this throughout life. In general, many energy groups are considered and these vary from 43 groups in the Dragon set to 164 groups using the General Atomic computational methods. By this technique the relative reaction rates in the system can be derived.

9. The plutonium 240 resonance has a very large flux depression within it, even in a homogeneous condition. Depending upon the precise conditions of the design this single resonance may hold down between 20 and 30 % in reactivity; it is obviously most important that a detailed treatment is given to this problem. There are two difficulties :

(i) the resonance is low lying, sufficient to be influenced by thermal neutron processes;

(ii) the resonance is broad relative to the maximum energy loss a neutron suffers following a collision with a carbon nucleus. This means that the technique which assumes that the resonance is narrow may be subject to significant error. It is noted that the Dragon project have considered this problem and refined their treatment accordingly.

10. Various techniques have been used for dealing with these problems and also for considering the effects of heterogeneity.

(iii) *Comparison with experiment.*

11. Although for U^{235} fuelling a great deal of experimental work has been completed, and, as a result, confidence in the methods of calculation has increased, I hope that at this session it will be possible to discuss the reliability and the uncertainties in our methods for Pu. fuelling when compared with relevant experiments, and also what our plans are for the future.

(iv) *Temperature Coefficients.*

12. With plutonium fuelling and graphite moderation, it is possible to have a positive temperature coefficient of reactivity associated with the moderator — indeed, this is a well-known characteristic of the Calder Hall system. This problem arises from the resonance of Pu^{239} at 0.3 ev. Calculations on HTR systems completed several years ago, indicated that there were design areas where a substantial negative temperature coefficient of reactivity could be found. In general, low graphite to Pu^{239} atomic ratios and low fertile contents produced systems with the most negative coefficients; so much so that it was concluded that temperature coefficient by itself was no real design problem. The low value of eta for the Pu^{239} resonance is most important since as the moderator temperature increases the value of eta for the core falls. The presence of the Pu^{240} resonance also provides a negative component to the coefficient.

13. We must note, however, at this point, an apparent disagreement between the paper by Massimo and that by Gratton. The calculations reported in the Massimo paper indicate a strongly negative coefficient over life, whereas in the paper by Gratton it is stated that calculations have indicated a less negative coefficient as lifetime proceeds. This is a subject requiring further examination to ensure that the conditions reached during the fuel cycle are acceptable from the tempeature coefficient point of view.

Fuel Cycles

14. The fuel cycle studies discussed in the paper fall under two main headings :

(1) those fuel cycles concerned essentially with once-through operation, i.e. starting with plutonium and a fertile material which may be either thorium or $uranium^{238}$, and to consider the performance of the first charge;

(2) those fuel cycles considering $uranium^{233}$ recycle with the system under equilibrium conditions. I shall deal with each in turn.

15. Several papers have indicated how the plutonium^{240} may be made to serve the dual role of fertile material and burnable poison. It has also been noted that because of the high absorption in this isotope, the permissible level of other fertile material is reduced.

16. The very high capture rate in Pu^{240} leads to the production of Pu^{241}, which is a superior fissile material to Pu^{239}. As a result of the removal of Pu^{240}, and production of Pu^{241}, reactivity is released which, under a homogeneous core condition, more than offsets the reactivity loss due to depletion and fission product build-up. The rate of release of reactivity may be adjusted by slowing down the rate at which Pu^{240} is destroyed. This is achieved by resonance shielding resulting from a more heterogeneous fuel arrangement. This can take two forms, either :

(i) making the fuel heterogeneous on a macroscopic scale by altering quite drastically the fuel element shape, or

(ii) selecting the size of fuel particle and achicving heterogeneity this way. The particle size considered, for example, in the Dragon paper was taken to be 500 μ diameter.

Whichever way is finally followed, it would seem to me that the effect of fuel heterogeneity is far more pronounced in a plutonium system than in an equivalent uranium 235 core. This will inevitably lead to close coupling of the physics and the material aspects of the design.

17. In the slide (taken from Fig. 6.1 of the Dragon paper) we see the effects of heterogeneity. In the first case, the core is considered homogeneous and we note a very rapid release of reactivity with the attendant control problem. As well as an engineering problem, the neutron economy suffers due to the loss of neutrons to the control material. Case 3 shows a much improved situation following heterogenising the system. Although we note that in this case the lifetime is reduced a little, the conditions considered in the UKAEA paper suggest that in certain cases an increased lifetime results from the more heterogeneous condition.

18. The ISPRA paper by Massimo draws attention to the very interesting spatial effects resulting from the type of reactivity characteristics we have just seen. In the slide (Fig. 16) we see the normal type of characteristic at the beginning of life. It is stated in the paper that at the end of life, due to the very rapid fall in fissile content the power in the centre of the core falls to the form shown (The maximum/average increases from 1.53 to 1.806). It is pointed out that by lumping the fuel and the addition of fertile material, a much improved situation may be achieved (as shown in the next slide, Fig. 17).

19. I do not propose to try and review the situation on fuel cost. As far as the comparison with the U^{235} system is concerned, I refer you to Table 6.1 of the Dragon report. If one assumes the same supply price for the core materials including fuel, the U^{238}/Pu fuel cycle is comparable with the U^{235}/Th system with the very important comment that the Pu system is working on a complete core replacement fuel cycle as opposed to a more complex 4-zone core. The worth of this simplified operation with Pu is worthy of note.

RECYCLES

20. Turning now to the question of uranium recycle, this is covered in the OAK Ridge and General Atomic papers. The Oak Ridge paper by Carlsmith and Thomas deals with the economic effects of recycling the spent fuel of a Pu/Th cycle. The comparisons are made between (i) no recycle at all; (ii) recycling the U^{233} only, and finally, recycling both the plutonium and the uranium 233.

21. From Table II of the paper it is indicated that recycling all residual fuel, results in a higher fuel cost, and that the most economic procedure is to recycle the U^{233} alone. The discarding of all residual fissile material leads to an increase in fuel cost over the best recycle case of 0.123 mills/kWh. It would be of interest to know the results of a calculation in which it is assumed that the thorium is segregated from the plutonium 239 and the U^{233} produced in one cycle is allowed to burn-up over a few cycles before being rejected.

22. The paper by Jaye *et al.* considers the long term fuel cycle capability of the HTGR, drawing particular attention to the possible use of low enrichment uranium. A concept is outlined in which ceramic uranium either as uranium oxide or uranium carbide is embedded within graphite fuel elements — it is envisaged that these ceramics would have a diameter of about 0.2″. This fuel lumping inevitably leads to thermal performance difficulties but, due to the shielding of the U^{238}, the required enrichment is less.

23. Section 3 of the paper on plutonium spiking is of particular interest to this session. An HTGR is considered under fuel cycle equilibrium conditions, with plutonium considered as the make-up fuel, and with U^{233} producing the majority of the power. Special attention is drawn to the power distribution problems following the insertion of a fresh plutonium assembly. It is shown that conversion factors between .78 and .85 can be obtained with a graphite system and figures as high as 0.95 are quoted with a fuel element containing a beryllia central spine.

24. It is of interest to note the low supply requirements for plutonium with these high conversion factors. In the case of the beryllia design, it is estimated that a feed of around 100 kg/year only is required to maintain a 1,000 MW(e) station in operation, i.e. from our plutonium feed we are extracting about 7,500 thermal MwD/Kgm.

25. For the reprocessing people, I should point out that all papers agree that the residual plutonium at the end of a charge is not worth reprocessing and can be discarded.

26. We are to note from the papers that the fuel cost estimates given depend on the assumed price of plutonium 239 but there are indications that for parity with uranium 235 fuel cycles approximately the same figure can be paid for Pu as U^{235}, the precise cost depending upon local conditions.

27. In conclusion, we must recognise that there is one other strong contender for the plutonium stocks that are being produced, and we must endeavour to make clear in our minds where the HTR stands relative to the Fast Breeder Reactor. The General Atomic paper looks at this point, and I am sure this is to be one of the non-technical problems to be solved in the future.

SESSION IV : DISCUSSION

Chairman : P. CAPRIOGLIO (Euratom)

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1. — AUTHORS' COMMENTS.

Dr. CAPRIOGLIO : I would like to make a comment on my own which is of a less technical nature than all those that have been raised by Dr. Gratton. It seems to me that in the field of thermal reactors, if one wants to burn plutonium, one finds that plutonium has to compete with the U^{235}. In this competition, it is quite obvious to note that natural uranium reactors are out of the question, because U^{235} is so cheap in natural uranium. It only costs 2 or 3 dollars a gram. It is almost certainly not worth taking the plutonium out of the spent fuel for that price. If one looks at slightly enriched uranium reactors, one finds that here plutonium has to compete with U^{235} that costs something like 7 or 8 dollars per gram and this starts to be in the range of interest for recycling without serious economic penalty.

So I think this is the main reason why there is so much sophisticated work carried out in the United States, to try and find out what the conditions are to recycle the plutonium in light water reactors. Of course, many uncertainties exist on the physics, on the fabrication cost, on the reprocessing charges. This is what makes it difficult, because there is only little margin.

When we come to highly enriched fuels, such as the ones we are talking about with the thorium cycle, here plutonium has to compete with very expensive material which costs 12 dollars a gram. Of course, the life is much easier. It seems to me this is the very reason why we are finding that in spite of all these economic penalties, metallurgical penalties and what not, the use of plutonium is more appealing in high temperature gas-cooled reactors, than it is on any other thermal reactor which is not making use of such an expensive fissile atom as the highly enriched U^{235}. This seems to me to be the fundamental reason for plutonium to look right in this reactor type and I would like to add this comment to what Dr. Gratton has just told us on many other aspects.

I would like to ask now the various authors to present their comments and their possible additions to what Dr. Gratton has just told us.

Dr. BRUNEDER *(Dragon Project)* : First I want to make another point to the comment of Mr. Caprioglio concerning the advantages of plutonium systems. The case which we have studied, was a so-called complete core replacement; you could get costs not much different under the same economic assumption from a more sophisticated multizone core in a U^{235} system. As you will see in fig. 6 (lower diagram) of the Dragon report 336, for a 4-zone core, assuming that the fissile amount is not recycled, we get costs in the order of 0.135 pence/kWh corresponding to 1.5 mills per kWh (p. 339).

For the U^{238}/plutonium system, which we found more advantageous than the corresponding thorium/plutonium system, we got 0.137 pence/kWh being equivalent to 1.55 mills/kWh. It seems that the physical reason why the complete core replacement in the plutonium system is better than the corresponding one in the uranium system, is that you can burn down your fissile material better. Using a complete core replacement philosophy gives in addition to the saving of money a saving in the capital cost, because the complete core replacement is probably cheaper than a 4-zone charge/discharge management.

Further I wanted to say that, owing to the programme we have used, the conversion definition does not include the destruction of plutonium-240 as fertile material. This will be now corrected and a very brief estimate will give conversion values which are about 4 % higher than those listed in the report.

Another point I wanted to make, is related to fig. 3.3.2 or 3.3.4 which shows the optimisation of the best reactor under certain economic conditions. As we heard yesterday, we are not quite clear whether the fabrication cost is really dependent on the heavy metal processed. People in our project have a different philosophy for the moment, which would more pronounce the uranium contribution towards the fabrication cost and you even could use this figure for reflecting this new philosophy, because here I have studied the plutonium prices as one parameter and the fabrication cost in terms of heavy metal as a second parameter.

For example : if the uranium component is twice as important as the thorium component, you just have to look up in this two dimensional diagram the optimum cost you get; so you can even reflect now the new philosophy with the old calculation we have done. If you look at this contour diagram, I want to stress that there are three things one wants to optimize :

— First (see fig. 3.3.1 and 3.3.3; one referring to the thorium, the other to the U^{238} system) in a complete core replacement with spent fuel discarded there is no intention to go to high conversion ratio, which is only interesting for crediting spent fuel.

— The second thing is assuming that you can neglect the fabrication cost that you go to the composition in the core where fifa is a maximum. If you have a dominance in the fabrication cost, you want to decrease your fertile amount, and these are the three independent positions and the optimum will lie in between these regions or boundaries according to the parameters you assume. This I have put together in fig. 3.3.2 and 3.3.4. Somebody might perhaps wonder why we have only studied the complete core replacement and not credited spent fuel. The argument is roughly the same as was given this morning : you have to start with the assumption that you do not have economic running reprocessing plants and hence you have to discard your fissile amount which is at the end available. Anyhow, compared with the U^{235} system, you will find that in the plutonium system you burn down much more of your plutonium.

— The third argument which I want to meet is why have I assumed 5,000 pounds for the plutonium price which is obviously more than offered on the market :

This is just to give comparison between the corresponding U^{235} systems and the plutonium systems. Assuming the standard fabrication charges as 150 pounds per kilo heavy metal, you break even at roughly 4,500 pounds for the U^{235}-thorium system.

The most important result is that as long as you regard complete core replacement discarding spent fuel, the U^{238} system is better than the corresponding thorium system. It has been mentioned by Dr. Gratton but I just want to stress again this point which is obviously not true if you assume equilibrium recycling.

Mr. CARLSMITH *(ORNL)* : I think that Dr. Gratton has given an excellent summary of the situation and I just have a few further minor comments to make. I would like to point out first, as is perhaps evident already, there is quite a wide variety of systems using plutonium that have been exhibited here. They range all the way from those that primarily burn plutonium and exhibit reactivity characteristics very much like partially enriched uranium systems, to those such as the plutonium-thorium with uranium recycle, which burn mostly U^{233} produced from the thorium and only use the plutonium to provide additional reactivity. These latter systems have quite high conversion ratios and resemble more the thorium/uranium-235 systems in their characteristics.

I would like to point out one thing which may not be clear in my paper that in the case in which we used a once-through system with plutonium/thorium, we assumed that the uranium which was produced would be sold and we assigned to that a value of 12 dollars per gram. Actually we have done other studies which indicate that we were somewhat conservative in this respect; the material which is produced here has a composition of about 98 % U^{233} and only 0.1 % U^{236}. Our value studies on uranium mixtures indicate that in a high temperature gas-cooled reactor, if we compare to a value of 12 dollars per gram for U^{235}, then U^{233} is worth approximately 18 dollars per gram and U^{236} is worth approximately minus 12 dollars per gram. This means that the price of a mixture resulting from irradiation depends very importantly on what the composition is. In the case that I am discussing here, the material would be worth almost 18 dollars per gram, whereas in some of the mixtures that were discussed this morning, where the U^{236} content was quite high, the resulting value of the fuel would be much lower.

I would also just like to mention a personal impression and that is that in all of these plutonium systems, the price at which plutonium can be purchased, is obviously important and perhaps the biggest thing that is needed before one can really consider plutonium seriously and start making plans for designing reactors to utilize it, is that a firmer price basis and a firmer basis of assured supply of the material needs to be established.

Mr. MASSIMO *(Euratom, Ispra)* : I would like to make a remark about temperature coefficients and its variation with life. These are only qualitative remarks as I do not have at this moment all the figures, but this can give some explanation

on my results on temperature coefficients. I fundamentally agree with the statement of Dr. Gratton.

Temperature coefficients can be split into moderator and fuel temperature coefficients. The fuel temperature coefficient is mainly influenced by the Doppler broadening in the plutonium 240 resonance and, as the spectrum softens with life, the absorption in plutonium 240 decreases and so does the absolute value of the fuel temperature coefficient and this appears from my paper. The moderator temperature coefficient is mainly influenced by the fissile isotopes of plutonium. As temperature increases, the f-value increases, because of the higher absorption in the 0.3 ev resonance, but at the same time the eta value is going to decrease. The overall effect will depend on the reactor composition. If f is low, the increase of f would be more important than the decrease of eta and the moderator temperature coefficient can be positive. But if f is already good (as it is the case in our reactor) this improvement can be insignificant compared with the decrease of eta and the temperature coefficient would be negative. Now during life, the spectrum softens so the absorption in plutonium 240 tend to decrease and, although there are the fission products, f might tend to increase (at least this may depend on the case). As a consequence, the moderator temperature coefficient can become more negative. In the average one will have no big variation, I think, of the temperature coefficient.

I think, another main feature of this reactor I have studied here, is that one can get a rather flat k-eff versus time curve so that one does not lose much into control. This can be obtained as a compromise between the S-value and the actual geometry. One wants to have the proper absorption in plutonium-240 (not too much and not too little) and so if the S-value is high, one will have few neutrons in the plutonium-240 resonance, so one will need little lumping; if the *S*-value is low, one will need a higher lumping.

Or course, I agree also with what Mr. Bruneder said. This is not the reactor which would be used for saving uranium ore, but it can be used either at the beginning if one has not got a reprocessing plant, or if, for some reason, one will have availability of plutonium fuel, either because there are no fast reactors to use it or because fast reactors have produced too much of it.

Mr. JAYE *(General Atomic)* : At first, when I realised that our paper was to be the last on Session IV, I was somewhat chagrined but now, after hearing the Session, I realise that our paper is almost a summary of the Session, so it fits in very well. Primarily we have tried to look at two things, the use of low enrichment as a primary fuel and the use of plutonium as a make-up fuel, and I stress this term make-up fuel with the idea in mind to change the HTGR physically as little as possible. With this in mind, I would like to point out, that when we use the plutonium, it really is a fuel cycle that does not differ very greatly from the ones that Dr. Stewart was describing this morning, with the exception that instead of using U-235, to make up the deficiency in the conversion ratio to 1, we used plutonium. We feel this is a good way for the HTGR to operate; we recognise fully well that the eta-

value of plutonium is considerably less than U-235 so we preferred to have as little of it as possible in the reactor from a fuel cycle standpoint, and this is the way we accomplished that particular aim. The other idea as to looking at low enrichment is simply the effect which Dr. Caprioglio mentioned, namely, the cheapness of uranium-235 as you get down below 5 % and certainly below 3 % of enrichment. When we did look at this, we found some difficulties in using the lowest enrichments, in that the lowest enrichments had a tendency to require large lumps. Large lumps had a difficulty to get high specific powers because of the thermal difficulties. We came to the conclusion that the use of low enrichment in the HTGR would probably require at least one of the following two items : that rather than oxide, we consider carbide and that perhaps the fuel ought to be cooled internally, e.g. in an equivalent large lump like one sees in an AGR type design where coolant flows around several smaller lumps. To the neutrons, it looks like a large lump; and to the coolant, it looks like a small lump.

I would like to say one other thing and that is that there was an oversight in the original version of our paper and a negation was left out in the very last line of the appendix. What looks like a positive statement ought to be a negative statement. This is not terribly important I admit, but it would bring some confusion. If I can quote it says "while this method deals with present worth accounting" and what it should say is "while this method does not deal with present worth accounting".

Now then as a fairly fast summary of the paper, I would just like to show a few of the figures that Dr. Gratton did not use. I wonder if I could have the first slide (fig. 2.1). In this particular figure, we have summarised what the fuel cycle costs would be in an HTGR fuelled with low enrichment as a function of the linear rating of the fuel lump and the cost of the ore. Now the rapid rise at about 2 kW per foot should be explained : we assumed that lumps below 0.2 inch would be impractical and therefore when the linear rating got below this value, we had to add additional stacks of fuel without making the lumps smaller. This led to lower specific powers and therefore higher cost. There is also the other effect that when you go to thinner fuel, you must go to higher enrichment and this tends to push it up but the first effect is by far the greater. We see in essence that if ratings of the order of 5 to 10 kW per foot are possible, then perhaps fuel cycle costs in the order of 0.7 to 0.8 mills/kWh with 5 dollars ore would be feasible under the economic parameters we listed in the report which I will not repeat.

In the next figure (fig. 3.4) we show the same data of fuel cycle costs in the HTGR with plutonium make-up (the upper figure is for an all graphite fuel element; the lower figure is for one that had beryllium spines). We see a minimum cost of between 0.8 and 0.9 mills/kWh for graphite and slightly above 1.0 for beryllium. One very interesting thing is on these figures : we have superimposed the dotted lines which are what we have termed the age peaking factor. If one uses a graded fuel cycle, then one inevitably has fuel of all ages mixed together. Superimposed on any gross flux distribution, you have a microscopic power distribution which is a ratio of the fission cross sections in essence of new and old fuel, and the numbers 1.2, 1.3,

1.4, 1.5 are the ratios of these fission reaction rates. You note therefore that the minimum costs are very steeply pointed towards the higher specific power cases, that is the cases with the largest value of carbon to thorium or beryllium to thorium ratio. However, these are not always achievable because the thermal design limits one to particular values of the age peaking factor. We generally think in terms of age peaking factors between 1.3 and 1.4 being compatible with our current fuel element design and the current maximum temperature allowed in coated fuel particles. That does lead to a rather strong limitation on just what minimum cost one could achieve in these fuel cycles.

Now if we could go on to the next figure, this is a summary of just everything that we were trying to achieve in this paper, and in this particular case, we have shown the sum of two parts of the costs, the depletion cost and the fuel working capital cost, as a function of the enrichment, and for the sake of comparison we have included the standard U^{235} make-up HTGR cycle. What we find here is that the low enrichment cases indeed show great promise, if one could use enrichments of the order of 1 % to 2 % which are from a physics standpoint feasible. However, we do find from a thermal standpoint, these are not feasible unless there is a significant improvement in the heat rating that one can get out of a big rod. The other thing that is of interest, is that in all of these cases U^{235} fuel, the plutonium fuel and practical low enrichment cycles seem to be within about 0.2 mills per kWh of each other on this ultimate goal which is merely the sum of these two costs which one cannot reduce.

Now, if we can just have the last slide (Fig. 1.3) : we have shown the uranium requirements which would be needed for HTGR complexes, which I will explain in just a moment, and fast breeder reactor complexes as a function of time. Now at the lower part of the curve, we have shown the installed capacity. (This by the way is the capacity which has recently been published by the Euratom Commission up until the year 2000, and we took the liberty to extrapolate linearly for the next 20 years.).

We have talked a great deal today about the difficulty of recycling fuel because of the large economy that was needed. It is rather interesting that in approximately 1973, we will have 10,000 electric MW and in approximately 1980 we will have 100,000 MW. We are talking indeed about very large economies in the not distant future, and I think from that standpoint it is valid to talk about costs in very large economies.

Now let me explain this use of the word "complex" for a moment : we all recognise that the fast reactor really requires plutonium in order to operate to its best extent. In this particular paper, we have been looking at HTGRs operating on plutonium feed. Let us assume that there is a plutonium producer which could either be a heavy water or organic cooled heavy water moderated reactor operating on very low enrichment or perhaps an HTGR operating on low enrichment. Then the plutonium is fed into a second HTGR which in this particular case has a beryllium to thorium ratio of 44 with a 4 year residence time. We see that in this particular case, the requirements will be roughly 2 million metric tons of uranium which will

have to be dug out of the ground. On the other hand if one takes the same heavy water reactor and loads the plutonium which is produced into a fast breeder reactor, then you get the range shown on the slide. I might just say that the range is due to looking at a fast breeder with a conversion ratio of 1.3, a second fast breeder with a conversion ratio of 1.4. It does indeed show that the conversion ratio is not the all-encompassing factor, which has also been said this morning. The specific power, by the way, in this particular fast breeder is some 700 kW per kilogram (systems specific power), so it is not a small specific power. It is interesting that the HTGR complexes have roughly the same uranium requirements to within, I think the uncertainties of the parameters used in the various reactors and the capacity requirements and I feel that this is an important point. It says that one might get as much mileage out of using the available plutonium in an HTGR, as he would using it in a fast breeder reactor unless one were to work and develop a fast breeder reactor which really meets all the goals that one sets, namely, conversion ratios of 1.5 or greater, and systems specific powers of 1,000 KW per kilogram and greater.

Dr. CAPRIOGLIO : As a layman, I am wondering why General Atomic people are so interested in extremely high conversion factors for fast breeders.

To come back to our discussion, I think there are a few important questions that have been raised by Dr. Gratton in his presentation and I would try to follow them one by one.

2. NUCLEAR DATA.

The first one was the question of accuracy of assessment and availability of calculation methods and nuclear data in the case of plutonium. We have found this morning that the situation was perhaps not completely satisfactory but good enough in the case of uranium and the problem is what is happening with plutonium, are we happy about what is available ?

Dr. SANDERS *(UKAEA, Winfrith)* : Because of the difficulties in dealing with the resonances in plutonium that Dr. Gratton has mentioned, it is important to establish the accuracy of methods of calculation, by comparing with sub-critical and critical experiments. At Winfrith, we have a programme of work which is aimed at testing the methods of calculation for plutonium fuelled systems. Some of this work covers the range of interest for the high temperature reactor and I would like to show you quickly some slides of results of near homogeneous sub-critical and critical experiments using plutonium fuel. The fuel that we have used for this work is plutonium-aluminium alloy containing 2 1/2 % of plutonium-240, so that the conclusions that we reach are essentially confined at the present moment to the plutonium 239 data and are not very sensitive to the plutonium-240. We first made some exponential experiments at a range of S-values from 2,400 up to 14,000 which spans the range of interest that has been covered in the papers presented in this Session. We have compared the results of these experiments with 40 group DSN calculations.

First Slide. — Graphite Moderated, Pu-Al Fuelled Assemblies

C/Pu	Measured Buckling metres^{-2}	Calculated k_{eff}	239/235 Fission Ratio	
			Measured	Calculated
14,520	13.24 ± 0.43	1.007 ± 0.014	1.33 ± 0.01	1.30
7,260 [a]	14.52 ± 0.41	0.995 ± 0.014	1.55 ± 0.01	1.45
7,260 [a]	15.56 ± 0.40	1.010 ± 0.013	1.52 ± 0.01	1.43
3,630	14.90 ± 0.41	1.011 ± 0.013	1.75 ± 0.02	1.57
2,420	13.48 ± 0.35	0.992 ± 0.014	1.79 ± 0.02	1.60

[a] Two assemblies at the 7,260 atom ratio were built with different fuel configuration to check heterogeneity calculations.

The first slide shows the results of the exponential experiment : on the left, we have the carbon to plutonium ratio and in the third column, we have the eigen value which is an indication of the accuracy of the calculation of the reactivity. You will see that the calculation of the reactivity is in fact within the limits of experimental error, over the range of compositions we have covered in these systems. I should explain that there is no other parasitic absorber in the system; they are just mixtures of plutonium and graphite. In addition, we have measured the ratio of the plutonium-239 to U^{235} fission cross sections, because these are sensitive to the neutron spectrum. These are given in the last two columns and you can see that in the more under moderated systems, there is some indication that we are not calculating this fission ratio correctly, which does indicate some error, either in our model for the thermalisation in graphite or in our nuclear data and group structure. Following on from the experimental experiments, we have made two critical loadings in the Zenith reactor, covering both ends of this carbon to plutonium atom ratio range.

The second slide compares the accuracy of the prediction of the reactivity, using a 43 group spectrum calculation and then a 6 group two dimension calculation to obtain the reactivity. The Zenith reactor, as you will probably know, is very similar in geometry to the Dragon reactor with important effects from the reflectors.

Second Slide. — Excess reactivities of zenith cores

Loading	S	Experiment	Calculation	Difference
Plutonium^{239}	2,670	3.8 ± 0.3 %	3.5 %	—0.3 ± 0.3 %
Uranium^{235}	1,070	5.3 ± 0.4 %	6.5 %	+1.2 ± 0.4 %
Plutonium^{239}	14,550	4.8 ± 0.4 %	6.3 %	+1.5 ± 0.4 %
Uranium^{235}	7,580	9.3 ± 0.8 %	10.4 %	+1.1 ± 0.8 %

We have also made in the past U^{235} loadings and you can see from this table, that we get comparable accuracy for prediction with the U^{235} loadings and the plutonium-239 loadings. We can normally predict the excess reactivity of these cores to between 1 % and 2 %.

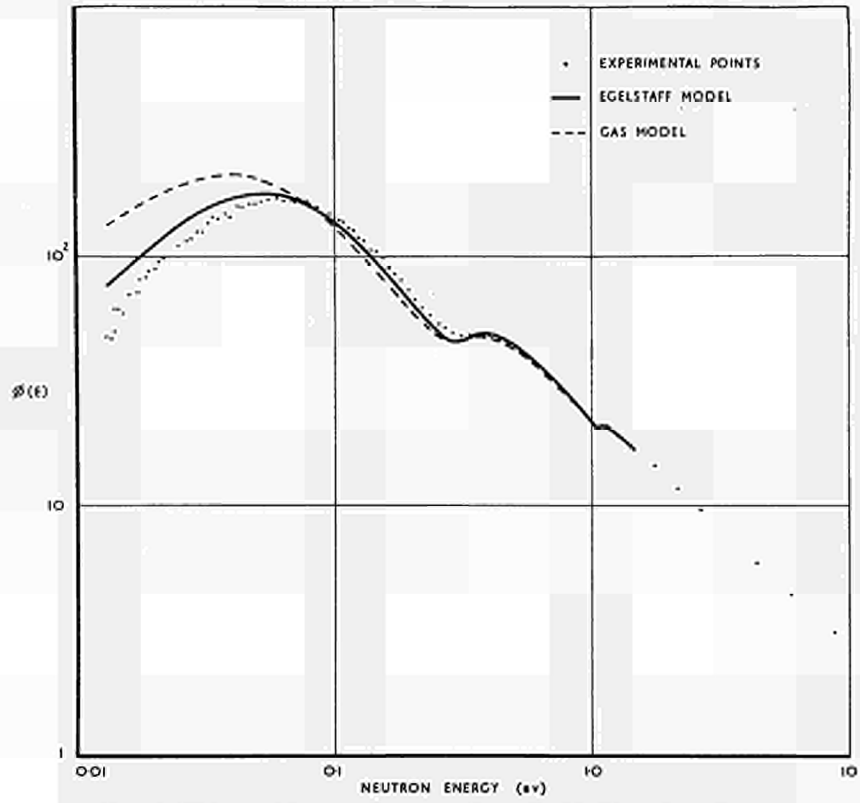
The third slide shows the temperature coefficient as measured on the Zenith reactor. Here we have a very small positive coefficient. You can see that the temperature coefficient of reactivity is predicted to within plus or minus 1 times 10^{-5} per degree Kelvin. Although the fractional error is large on the prediction, this is a satisfactory absolute accuracy for practical purposes. The small net coefficient is in fact a balance between various effects that were mentioned by Dr. Gratton, the changing effects of leakage, the changing eta and the changing value of *f*. These values are for the core with an S-value of 2,660.

Third Slide. — Temperature coefficients of reactivity Zenith plutonium core
(Units of 10^{-5}/°K)

Region	Temperature range	Experiment	Calculation
Core Plus End Reflectors	300-500° K 500-700° K	$+0.1 \pm 0.2$ $+0.8 \pm 0.2$	$+0.8 \pm 0.2$ $+0.6 \pm 0.2$
Side Reflector	300-500° K 500-700° K	$+4.4 \pm 0.3$ $+3.0 \pm 0.3$	$+3.6$ $+2.2$

The other result I would like to show you in the fourth slide is a measurement of the neutron energy spectrum in the Zenith reactor. Because of the sensitivity of the plutonium cross sections to the spectrum, it is important to check our calculations of the energy spectrum and we have done this by means of a neutron time of flight experiment from the Zenith core. You can see the effect of the plutonium-239 resonance at 0.3 eV and, with the eye of faith, the effect of plutonium-240 resonance at 1 eV. The dashed curve is the spectrum calcultaled using a free gas model for the graphite, which as one expects gives a too soft a neutron spectrum, while the full line is the spectrum calculated using the Egelstaff model for graphite which allows for crystalline binding which, although not a perfect fit, certainly does much better than the gas model. Although obviously one can refine this a bit further, we feel that the Egelstaff model predictions are good enough.

Our general conclusions from these studies are that for near homogenous systems fuelled with plutonium-239, our methods of calculation are adequate for practical purposes. We are currently extending these experiments to fuels containing up to 26 % plutonium-240, in order to test more stringently the calculations of the plutonium-240 resonance capture, which has such an important effect on the lifetime characteristics.



Fourth Slide. — Neutron Spectrum from Zenith Plutonium Core.

Mr. CARLSMITH *(ORNL)* : I would just like to ask Dr. Sanders whether these experiments are written up in the form of reports that are available?

Dr. SANDERS *(UKAEA, Winfrith)* : The experiments on the exponential systems have been published now as report No AEEW-R347.

Dr. CAPRIOGLIO : I think that it looks quite encouraging to see how the experimental values fit with calculations. I am wondering if reactor designers feel satisfied with this.

Dr. STEWART *(General Atomic)* : I would like to make a comment, both on remarks that were made this morning and this afternoon. It appears from things that have been presented, that the cross section data are indeed in good shape and there have been many boasts, particularly by us, that we can calculate things quite well. I would like to caution everyone though that we do our best job of calculating experiments after the experiments are done. This is a somewhat cynical statement but nevertheless somewhat true.

So we do have sufficient confidence in experimental data from the point of view that we can do a good job of assessment of performance of reactors. However,

I do not want to leave the impression that we can calculate absolute reactivities at the beginning of life of any reactor with great precision. But again, we think, we do have good enough data such that, once a reactor has been loaded and operated, we can make adjustments in the thorium loadings or fuels loadings of subsequent reactors and get very nearly the kind of operational characteristics that we would anticipate with our current data.

Dr. CAPRIOGLIO : I think this summarizes quite well the situation and I think we can then go to the next item on our agenda if there are no more comments.

3. — EFFECT OF PLUTONIUM ISOTOPIC CONCENTRATION.

I would like to raise now the question of effect of plutonium isotopic concentration which is a very important one to my mind, since obviously in the United States there is a tendency to consider by plutonium, what comes out of light water reactors. On the contrary in Europe, and in particular in the United Kingdom, there is a tendency of considering plutonium as it comes out of natural uranium gas-cooled reactors. So I think it is in order to clarify what the importance of this difference is. One obvious difference is the content of plutonium-240 so that perhaps the importance of lumping would change with the variation of 240 concentration, but I can be wrong here. I would like to invite comments on this.

Dr. GRATTON *(UKAEA, Winfrith)* : When we formed our initial calculations on plutonium fuel cycles, using plutonium not as a fertile material but as a burnable poison, we showed how by careful adjustement of the fuel element, we could get a near ideal reactivity time characteristic. But the question quickly came back : if you require to tune your reactor to this particular level of plutonium-240, what would happen if you changed over from Magnox plutonium to AGR plutonium which has a higher plutonium-240 content. Of course, the immediate comment was : it all depends how much the AGR plutonium costs. If you assume the same price, you are paying rather expensively for a lot of plutonium-240. But, as was shown in the Dragon paper, a plutonium reactor designed this way, has a degree of freedom in the design, which the U^{235} system does not have, namely, the choice of particle size. We recall seeing a reactor with a rising reactivity time characteristic in the homogeneous case, virtually a flat characteristic in the heterogeneous case. I feel that as far as the change from Magnox plutonium to AGR plutonium is concerned, it should be possible to move one's design to satisfactory conditions by some small change in the range of particle sizes you are constructing your fuel element on.

Dr. CAPRIOGLIO : Could I ask Dr. Gratton what *range of particle sizes* you are really talking about? Is it a wide range or a relatively narrow one?

Dr. GRATTON *(UKAEA, Winfrith)* : It is very difficult yo give precise figures on this. If you go for a very small particle size, then you take your heterogeneous effect from fuel element shape. But I would have thought that a range from say 200 microns to 500 microns would make a world of difference to the characteristics you obtain from your reactor.

Mr. FISCHER *(General Atomic)* : I would like to make just a quick comment on the effect of plutonium-240 on the temperature coefficient.

We have done some calculations for the low enrichement system and found that the effect of plutonium-240, not due to Doppler broadening but due to spectrum changes, is very effective in changing the temperature coefficient. There might be a very slight shift in spectrum up in the 1 eV range, but there is enough up-scattering at high temperatures, so that the plutonium-240 contribution absolutely dominates the temperature coefficient. Not the Doppler broadening but just the effect that you scatter more neutrons up into the higher energy range. And obviously since plutonium-240 is a poison, it is a very welcome negative contribution which gets stronger the hotter the core gets, which is a very nice characteristic too. Obviously we observe at the lower temperature range the same effect as you did with the plutonium-239, which can, depending on the conditions, add positively to the temperature coefficient. It can also add negatively, depending on what fuel concentration, what S-value you have and what temperature you are at.

Dr. GRATTON *(UKAEA, Winfrith)* : In fact, I agree with every word you say : the 240 does have these two temperature coefficient effects with it. I am sure the thermal spectrum effect is an important one and I think, I said the very fact that it is there leads to further complication of your calculation. But we must bear in mind that in these systems the plutonium-240 concentration does fall considerably over life and the component that you are referring to, will tend to fall off, but I think this is very system dependent.

Mr. FISCHER *(General Atomic)* : I would just mention that in the Peach Bottom reactor which operates with 93 % U^{235} enrichment, the tiny amount of plutonium-240 produced in the life of that reactor is a major contribution to the coefficient; so it takes very little plutonium-240, depending on the system, to significantly affect the temperature coefficient. We were amazed ourselves when we first did the correct calculation of how important the Pu^{240} is, even in a highly enriched uranium/thorium system.

Dr. SHEPHERD *(Dragon Project)* : It is just a matter of obtaining a little clarification on this question of self-shielding by going to large particle size. I did not find in his paper any reference to the constitution of the particle which is obviously rather important if you are specifying self-shielding. I wonder, if Mr. Bruneder would like to comment on that and indicate what is meant by 500 microns.

Dr. BRUNEDER *(Dragon Project)* : The 500 microns is the kernel size.

Mr. HAUBERT *(Dragon Project)* : I should like to add that these particles are considered to be plutonium di-carbide mixed with a certain amount of U^{238} or thorium, and that the density of this kernel was considered to be 8 grams per cm^3. I should like to add also that these calculations are not done in order to get very accurate results, but only to have an indicative value and to see what could be the influence of the particle size and composition on the fuel burn-up and performance.

Mr. FORBES-GOWER *(CEGB)* : I believe, you were asking for clarification on the isotopic concentrations used and I am not sure that you received much of an answer on it. I would like to throw the cat among the pidgeons, by introducing some new data which the Authority will be particularly displeased at; that is that the Board are increasing the irradiation discharge limit in the Magnox reactors and if you consider the figures from which the earlier numbers were obtained, they came from Rose and Syrelt of 1959, using the same graphs, you would get a new concentration of approximately 76 % plutonium-239, 17 % Pu^{240}, 5 % Pu^{241} and about 2 % Pu^{242}.

Now I would like to ask if anyone has done any re-evaluation of these same curves, since these were almost certainly from Paper-80 and our present experience in the Board is that long term reactivity calculations are a long way out using Paper 80. We ourselves have not yet got round to publishing new figures using the later Argosy method, but I would like to know if anyone has re-evaluated what the isotopic concentrations in plutonium from the reactors producing it in the United Kingdom would be.

Dr. CAPRIOGLIO : It looks as though there is no specialist from the UKAEA who is wishing to answer.

Dr. SCHROEDER *(Dragon Project)* : I think if we would use these figures in the type of cycles that Dr. Bruneder and Haubert have described in their paper, we would find no difference using this composition of plutonium compared with that we have used in the paper.

Mr. FORBES-GOWER *(CEGB)* : I think I would agree. However, I was interested to see that the figures are very similar to the General Atomic's figures and therefore comparison with that would still be valid. But the point that I would like to bring particularly out of this, is that the reason, or one of the postulated reasons, why the reactivity build-up in our Magnox reactors is wrong, is because we are not predicting the right amount of plutonium production. Since the reason for this being wrong is bound to be connected with the isotopic conversion from 238, our methods of calculating the relative amounts of 240 and 241 will be no better and perhaps even worse, since the data certainly would not be any better known than for 238 which we have had for a lot longer. In which case our whole set-up of ratios may be altogether wrong and I would like to know if anyone has made any attempts using multi-group methods to repredict the plutonium content in irradiated Magnox fuel?

Dr. CAPRIOGLIO : Is there any other comment on this question of effect of isotopic concentration?

Dr. GRATTON *(UKAEA, Winfrith)* : I tried to answer this question from the point of view of this burnable poison idea with 240. I wonder if the General Atomic people have looked at the effect of varying the isotopic concentrations on the recycle case and whether it affects the answer significantly.

Mr. JAYE *(General Atomic)* : I can comment really only very briefly on this. We have done this work extensively, but not in the graphite moderated HTGR, but rather in the advanced beryllium oxide project. In that particular system, we were looking at fuelling very much like that which Mr. Massimo talked about, namely, very low reactivity swings. We indeed found that, as one would interpret Mr. Massimo's results, varying the 240 concentration or alternatively varying the lumping for a given concentration, changes the reactivity variation with time very markedly. From this standpoint, I can fully sympathise with the gentleman from CEGB in that a small error in the alpha of 239 really could result in a significant change in the 240 content which would look like an enormous poison in such a reactor. I am not at all surprised that he has a certain amount of difficulty.

Mr. MASSIMO *(Euratom, Ispra)* : If one compares my result with Bruneder's result, one can see that going from a 18.8 % plutonium 240 to 16 %, fifa in this type of cycles will slightly increase.

Mr. BLOMSTRAND *(Dragon Project)* : I should like to make a comment on what Mr. Jaye was just saying, that a small change in the eta value of the 239 isotope can make quite a great difference in the results. It has been mentioned that the eta value for plutonium-239 is very spectrum sensitive, so that I think would be possible to design a reactor which would keep the graphite as cold as possible, this could improve the burn-up performance of these systems, because it would improve the eta value for plutonium-239.

I should also like to enquire whether there are any accurate data for the energy dependence of the eta value of the plutonium-241.

Mr. JAYE *(General Atomic)* : I agree with Mr. Blomstrand that the eta value is not very well known for Pu^{241}, however I do not entirely agree with him on the question of temperature. When I have looked carfully at the spectra, I find that the S-value has a much stronger effect on the spectrum than does the temperature and that if one wanted to get very high eta-values for 239, the way to do is to go to extremely high S-values. One of the things that is most desirable on the plutonium cycle is the achievement of a very flat reactivity. If one does do something to improve the eta-value for 239, you absolutely defeat that purpose, because you push all the neutrons out of the 240 resonance and it does not look much like a burnable poison at that point.

Mr. FORBES-GOWER *(CEGB)* : I wonder if I could just verify that. Some five years ago, I did some work using a multi-group programme and we did repeat the runs at 1,000° K moderator temperature and at 700° K moderator temperature and we have one at 500° K. The difference in reactivity lifetime as a result of decreasing the temperature, is to result in a shortening of lifetime of about 50 MW days per fissile kilogram. The reason is not the change in eta but, as the last speaker mentioned, it is because you have taken the flux away from the 240. I think that those who have not worked much with plutonium will hardly appreciate how impor-

tant it is that the 240 must be converted, so that it acts both as a burnable poison and as a fertile material. Nearly always every step must be made to obtain high absorption in 240.

4. — FABRICATION.

Dr. CAPRIOGLIO : I suggest we go a step further now and look at one that seems to me an essential aspect of using plutonium in a high temperature reactor or indeed in any reactor, which is how much it is going to cost to make the fuel with plutonium. This is not any more strictly a problem of physicists and this would make therefore a nice change for a moment to our attention. I would like to ask Mr. Carlsmith or Mr. Lotts if they would like to make some comments about what is happening in Oak Ridge in this field.

Mr. LOTTS *(ORNL)* : I do not think I could really comment, except to say: in the paper which we had in the second Session, there are some costs given and one could infer from that the penalties which would be associated with the fabrication costs.

Mr. JAYE *(General Atomic)* : I would like to pose another question to Mr. Lotts, if I may, along the same lines, namely : What do you feel the difference is in fabrication if one is considering recycled uranium or alternatively plutonium with or without recycled uranium ?

Mr. LOTTS *(ORNL)* : I think I got the combination : the first one is just plutonium, plutonium/U^{233} and U^{233}. I think that again you would have to take into account the amount of shielding which is required in the case of the U^{233}, the percentage of U^{233} and plutonium that you would have. So again you could refer to the curve which gives the penalty for the shielding.

Dr. SCHROEDER *(Dragon Project)* : I noticed that during the Session yesterday there were investigations on how to produce plutonium/thorium particles and I would like to ask for what purpose or for what type of fuel cycle this particle is envisaged.

Mr. CARLSMITH *(ORNL)* : I am not really sure I understand the meaning of the question. The question concerned the purpose of having a particle with plutonium and thorium. I think the calculations that we have presented in our paper indicate that one can get quite good fuel cycle costs in a homogeneous system that utilizes the plutonium/thorium fuel if the plutonium can be obtained at a reasonable price, say something less than the cost of fully enriched uranium.

Dr. SCHROEDER *(Dragon Project)* : I should add that I have understood and read your paper, but I think you are speaking about reprocessing this type of fuel. If I understand you right, you will then have, as a result of the reprocessing, a mixture of plutonium and uranium, so, except at the beginning of initiating this type

of cycle, there would never arise an occasion to produce pure plutonium/thorium particle.

Mr. DOUGLAS *(ORNL)* : Perhaps the confusion on this particular point arises because Mr. Nicholson was really expounding on the flexibility of the Sol-gel process and its capability to make a variety of different kinds of fuels. He was mentioning the fact that pure plutonia had been made in microspheres using this process and that as an experiment, we had mixed oxides of plutonia/urania and plutonia/thoria with, in this particular case, no other point in mind other than to discover if such fuel could be made. His point was not directed towards any immediate application.

Dr. SHEPHERD *(Dragon Project)*: I want to deal with a more general point which is rather important. From all the papers we have seen today, it is quite obvious that we are reaching a state of affairs where there is such a proliferation of fuel that the job of developing and testing it, is becoming rather a difficulty. I have a feeling we ought to do something about standardization. We have already come to this conclusion in Dragon with respect to plutonium fuel. We decided that we should not go ahead and investigate plutonium/thorium, but we would assume that these materials would be segregated. I am wondering if we should not turn in that direction in all cases, because otherwise we are going to find ourselves coming to fabricate and test an enormous range of different types of fuel. Should we not just for practical reasons, apart from the scientific reasons, consider segregation very seriously to enable us to reduce the amount of development needed.

Dr. CAPRIOGLIO : Am I right in assuming that this means in fact diluting the plutonium as well, because if one wants to have sensible burn-ups within the metallurgical limits of coated particles, one is likely to need dilution. What kind of dilution has been thought of at Dragon and what in other places.

Mr. HUDDLE *(Dragon Project)* : We faced this problem some two years ago, when we started to think about this and as a result we have concentrated our work on what we call the "UC 10" type fuel. The "UC 10" does not imply precisely 10 atoms of carbon. For example, the one that we are putting in the reactor is about U/C = 1/11. It is essentially a particle where the fissile material either plutonium or uranium-235 or 233 is dispersed in the particle within a matrix of carbon. We think such a particle should be capable of virtually 100 % burn-up and so far the results we have obtained are most promising. We, in Dragon, think such a fuel has good prospects.

Dr. CAPRIOGLIO : Could I ask the Oak Ridge people about their ideas about the segregation of plutonium and dilution of plutonium in coated particles? Well, if you do not want to comment, I would ask the same question to General Atomic, if any thought has been given to this problem.

Dr. STEWART *(General Atomic)* : Some thought has been given to it, but I guess well not enough to make really any intelligent comments.

Dr. CAPRIOGLIO : Any comment on the main suggestion of Dr. Shepherd that some sort of standardization is now needed and that the suggestion to start with segregated fuel is perhaps a wise one?

Mr. CARLSMITH *(ORNL)* : Standardization is certainly desirable, but it sounds to me from the comments in the last few minutes, that the group here does not really know enough about what system we should standardize on. I think, we are still in an exploratory stage in this area and we are trying too find out what makes sense. That is why we are looking now at a variety of systems, not that we are actually proposing that all these systems that we are looking at, be built.

Dr. SHEPHERD *(Dragon Project)* : My proposal was a simple one, that was to separate each particular fissile or fertile element in the particles, giving thereby a free choice about the nature of fuel constitution by mixing the particles in any desired ratio.

Mr. JAYE *(General Atomic)* : The only comment I would like to make, which is mostly a comment on Mr. Carlsmith's remarks, is that I think unless we do something along the line that Dr. Shepherd is expressing, it is going to be very difficult for us to figure out what makes sense. Because I think what makes sense will inevitably be determined by what makes economic sense in the fabrication procedure, and if it should turn out that plutonium is much harder or much easier to deal with than U^{233}, we must learn this before we can decide whether plutonium is much better or much worse than U^{233}.

Mr. BLOMSTRAND *(Dragon Project)* : I would like just to draw the attention towards what Mr. Gratton was saying in his introductory paper, namely, that he would be interested to see the results of an analysis of the system in which plutonium went through rather quickly and the thorium was in the system for a longer time. I do not want to stick my neck out on such a system, but it is quite likely that this is the best way of utilizing plutonium in the system. This will require separate plutonium and separate thorium particles.

Dr. BILDSTEIN *(SCAE)* : I would just like to underline Dr. Shepherd's statement that it is advisable to work on segregated fuel, because you not only have the benefit to work on a less number of varieties, but in addition in producing large amounts of thorium carbide particles, you can reduce the costs of production quite drastically. Because in this case, you do not have the necessity to work under critically controlled conditions. You can easily coat thorium carbide particles for instance in a vacuum coating furnace up to a rate of 10 kg per batch, which will reduce the costs by a factor of 2 or 3 approximately.

Dr. SCHROEDER *(Dragon Project)* : Just one further comment to expand on that what Mr. Blomstrand just said. We have thought about this plutonium segregation business in the Dragon Project. As mentioned already, we could not find any reason why the plutonium should not be segregated. Because you would in

in all circumstances use the plutonium in this type of reactor preferably to make up for the deficiency of neutrons, to use it as a seed material. In that respect, you would not reprocess the plutonium and for that reason you would not like to put that plutonium through the reprocessing plant and discard it afterwards, but would rather like to discard it together with its fission products. This is one further reason why we should really just produce a plutonium particle.

Mr. JAYE *(General Atomic)* : I should like to ask Mr. Blomstrand to clarify just one point for me. This morning I was confused on one point concerning feed and breed, and I want be sure I am not confused on the same point here. When the thorium is inserted into this cycle, you are talking about, does it have any fuel with it?

Mr. BLOMSTRAND *(Dragon Project)* : It is not really a physics problem but a heat transfer problem. From a fuel cycle point of view I would not like to have any fuel in the thorium, but if somebody wants to prime it with something for the heat transfer point of view, I think this would be all right still. I do not know if anybody else wants to expand on the matter.

Dr. CAPRIOGLIO : So the point is that one likes to have a rather constant rating in the breeder elements, so that one has to start with some fuel in it, so that there is a certain production of heat in them and then going with a conversion factor of 1 in the breeder zone, so that the production of heat is somehow constant during the life of the reactor. I think this I understand.

Mr. HOSEGOOD *(Dragon Project)* : I think, the chairman has summed it up very well in the context of the type of reactor in which we are interested at the moment in Dragon. The idea is to have a fixed breed which remains in the reactor without any fuel movement over a very long period of time. In this context, there are very clear advantages in maintaining a constant power output from this breed region. However, it is equally possible to envisage a reactor with a movable breed region, that is one which is fed relatively frequently and with some discharge relatively frequently. In such a case, one could envisage systems in which the breed region feed would be plain thorium elements and the breed power as a whole would be maintained constant by relatively frequent discharge of some breed elements and their replacement with new ones. This would mean that one was losing some of the advantage of the breed and feed type of system by discharging from the system breed elements containing uranium-233. This uranium-233 would of course have to be recovered by reprocessing, so such a system would not be of any interest, unless reprocessing costs were attractively low.

Dr. CAPRIOGLIO : I think we have gone through quite a lot of questions concerning plutonium. I do not try to summarize but is seems at least obvious to me that plutonium looks very attractive in these high temperature systems and that it seems

unlikely that very serious snags are going to appear, unless fabrication costs are going to be too high. But fabrication costs are already so high with uranium that I cannot really believe, they could become sensibly worse. In other words, we already have to deal with glove boxes, with controlled atmosphere, with all sort of equipment that people usually think of, when they think of plutonium. So I cannot believe that the fact of using plutonium is going to make such an enormous difference in fabrication costs as to overcome all the advantages we have been talking about this afternoon.

Dr. BRUNEDER *(Dragon Project)* : I just wanted to confirm your statement that the fabrication cost is not too important when you look in the Dragon report fig. 3.3.4. You have here the variation of the optimum fuel cost, changing the fissile cost and the heavy metal fabrication cost. You see that the uncertainty in the fissile amount has much greater influence than the variation in the fabrication cost which anyhow at 150 pounds is quite high. If you would make it, let us say 30 % more expensive, it would very insignificantly increase the optimum fuel cost.

Dr. CAPRIOGLIO : Just one minor remark I wanted to add is, that if it is true that segregation is useful and it is well worth doing, then I think the exponents of the oxide root should think a bit about how to dilute plutonium oxide particles. The carbide is easy, because it is relatively easy to add carbon. I do not think one can add oxygen in the same way, so one has to add something else. I would be very much interested to see people doing work on plutonium oxide diluted with something else, which looks all right both from the neutronic and the metallurgical point of view.

5. — UTILIZATION OF RESOURCES.

I do not think, we should go any further now, and I would like to ask for comments on Dr. Stewart's paper this morning. I believe everybody has it in his mind in spite of this long interruption of dicussion and I would like to ask for comments.

Mr. HOSEGOOD *(Dragon Project)* : I remember joining enthusiastically in the applause at the end of Dr. Stewart's paper this morning. But I have since then been thinking a little further and I have recovered just a little from being carried away. The theme of Dr. Stewart's paper was largely concerned with utilization of resources in relation to the cost of recovery of ores. He did draw our attention to a very valid point, which is sometimes overlooked, and that is that there is plenty of uranium; it is largely a question of how much it costs to recover it.

The utilization of resources is the right way to look at it, with some economics in it, rather than merely adopting a pious expression and saying, because I have a high conversion ratio, I am saving the world from running out of fuel, regardless of cost. However, in terms of this ore utilisation, I would like to know what recovery of uranium-235 from natural uranium in a diffusion plant was assumed. I gathered an impression from I must admit a rather superficial perusal of the paper, that it was assumed that if you wanted 7 grams of uranium-235, you just dug up a kilogram

of natural uranium and put it through the diffusion plant. I am sure that this cannot be so and if there are no military secrets involved, I would like to know what fraction actually was employed here.

There is also a point, which perhaps is completely unimportant, but again I am ignorant of the answer and would like to know what it is. Concerning the energy conversion efficiency, Dr. Stewart did in fact mention that the high thermal efficiency of the high temperature gas-cooled reactor did result in a large net output of electricity for every gram of fuel burnt. However, from this we must subtract some small amount to run the diffusion plant to supply another gram of fissile material. In a very rapidly expanding power programme, one can envisage some sort of nightmare world in which the rate of commissioning high temperature gas-cooled reactors, can just keep pace with the demands of the new diffusion plants to fuel the next generation.

Turning more directly to costs, we have to allow something for the cost of building diffusion plants in the far future, when we envisage that the rate of installation of reactors is out-stripping the capacity of existing plants. Now if one is comparing high temperature gas-cooled reactor systems, with comparatively modest conversion ratios on the one hand, with a programme of natural uranium plus fast breeder reactors in the context of a very large and expanding power programme, one tends to congratulate oneself with the HTR in having a low capital cost system. However, strictly speaking, each HTGR should carry its own share of the capital cost of the new diffusion plant one is building and commissioning. At present, we take this into account by the present day price of U^{235} which we assume is already paying interest on the diffusion plant. Now it is not very clear whether the interest charges are included in the price of U^{235} or whether the capital cost of the existing plants is assumed to have been written off as military expenditure a long time ago. If one did in fact build a diffusion plant today and demanded a reasonable interest rate on the capital, as well as all the usual running costs and other expenses, would the price of U^{235} be greater, the same or less then the 12 dollars a gram which we assume today?

Dr. STEWART *(General Atomic)* : On the first question, namely, what did we assume in the way of U^{235} yield per gram or uranium mined : We assumed that the diffusion plants would be operated as they are currently being operated. As I recall it, the U^{235} in the tails from the plant amount to 0.25 %. The amount of uranium required for 1 gram of U^{235}, is then, something like 200 grams of uranium for each gram of U^{235}. Perhaps Mr. Carlsmith can check me on this. Is that about right?

Mr. CARLSMITH *(ORNL)* : Yes, it is very close to that.

Dr. STEWART *(General Atomic)* : On your second question, which dealt with the adequacy of diffusion plants to support an economy consisting of enriched uranium reactors : This problem, of course, exists for any enriched uranium reactor that one might assume. First, before going further, let me comment that this again points

up the importance of designing reactors that require the smallest possible inventories of U^{235} and the smallest net consumption of U^{235}, so that aside from the possible use of natural uranium reactors, I think this makes even a stronger case for choosing reactors during this fast build-up period to minimize the uranium requirements.

We have looked at various combinations of reactors, what we call symbiotic systems, and these are discussed in the paper. A symbiotic system, we define as a system of reactors consisting of one reactor which generates fuel that can be used in a second reactor. The second reactor may be the same reactor or a different one. Examples of symbiotic systems would be for example, the magnox reactor and a fast spectrum reactor, or a light water reactor and a fast spectrum reactor, or an HTGR starting with U^{235} and an HTGR using recycled uranium. It has been our conclusion in our examination of symbiotic systems, that the amount of plutonium one can produce in thermal spectrum, low enrichment or natural uranium reactors, is quite small in terms of the plutonium requirements that are demanded by a fast spectrum reactor economy. As a result, if you look at the curves in our report, you will see that it certainly is not justifiable to build thermal spectrum reactors, solely for the purpose of producing plutonium to start up fast spectrum reactors. Indeed both from a resource conservation point of view and an economic point of view, one would be better off to start fast spectrum reactors up with U^{235} and use the plutonium that they generate in subsequent fast spectrum reactors.

On your last question which had to do with the cost of U^{235} in new diffusion plants, I think there is a good deal of mystery surrounding this, because of the secrecy that has been imposed on the diffusion plants in the United States and probably elsewhere. But there was a report, a few months ago by the Ebasco Corporation in the United States (see "Nucleonics", vol. 22, nr. 9, page 19, September 1964) which was an unclassified examination of the probable cost of separating U^{235} in plants to be built independent of the Government. Their general conclusion, as I recall it, was that the U^{235} could be separated for approximately the same cost that the AEC now establishes for U^{235}, provided the volume of business were about the same as in the past.

Dr. CAPRIOGLIO : The more I listen to you, the more I feel embarrassed because Mr. De Bacci and myself are writing an article which does not include many new things with respect to what we are just saying. You have just to add a couple of things and we would just avoid writing it at all.

There is one symbiotic system that you have not considered and which could be perhaps interesting to look at; it is the symbiosis of a fast reactor producing plutonium which is going to be burnt in an HTGR. This seems absurd at first sight, but there are good reasons for taking it seriously.

Mr. FORBES-GOWER *(CEGB)* : Could I ask Dr. Stewart and apparently yourself also a bit more about this U^{235} cost. The figures published by the Americans are very nice and a great amount of our economic studies have been based on the published U^{235} price. However, they have not been challenged by being bought

by a competitor. Is the price somewhat nice so as to encourage people to buy American reactors and American fuel, and would the same price be offered to a competitor?

You say that a second study has been done to start a plant today to make U^{235}, so the issue would appear to be avoided by this remark. In this plant, what was the cost of the electricity used for the process? We have understood that one of the reasons for the American plant cost being less than that of the United Kingdom plant, is related to the cost of electricity as supplied to the plant. In the Ebasco Report, I wonder if it were the typical conventional electricity price that was used, or if special arrangements were made between the producer of electricity and the U^{235} producer, on the understanding the U^{235} was being made for him. In which case, he might be prepared to let the electricity go a lot cheaper. Now I could give examples that our costs would be altered by approximately 30 % for U^{235} if such an arrangement were entered into. But the Board cannot consider entering into such an arrangement, if it is not the Board that is getting the U^{235}.

In this connection I would like to return to the question on the cost of plutonium. If we are considering a symbiotic system, there is surely no point in costing the reprocessed fuel according to the fissile component of it. The only cost is that of fabrication, in which case it is artificial to give plutonium any price, if it is being used again by the producer. If the producer is also the consumer, there is no point in costing plutonium. Now the systems discussed today have all started with the plutonium cost. It was thought by the proposers of these systems that somebody who does not produce plutonium is going to be able to buy plutonium from someone else (other than that which they later make themselves). If this is the case, I think it might be a little naive to assume the low prices fixed for plutonium in their calculations. If, for instance, the Board were being asked for plutonium by someone else, they would have to debit their fast reactor programme with this loss of plutonium. Therefore the cost would have to be evaluated on how much it would affect the fast reactor programme, not how much it was to be compared with U^{235} in some other system. I also wonder, to how many nations or countries would any producer of plutonium be prepared to supply enough plutonium to make an atomic bomb?

Dr. CAPRIOGLIO : I think, there are many questions raised by your remarks. I do not think, we can profitably take up and discuss here the old polemic about how much it costs to enrich uranium. Because, first of all, I do not think there are really many people expert in this field in this audience and furthermore, because I do not feel we will ever get a satisfactory answer. On the other hand a few other questions have been raised which I think are worth taking up. Is Dr. Stewart wishing to answer to some of them?

Dr. STEWART *(General Atomic)* : On your questions about availability of U^{235} at current costs, I am afraid I really cannot comment any more than what you would read in the technical journals and so forth. On the basis for the cost of diffusion,

I cannot supply any more information than that which I previously quoted from memory as what I read in "Nucleonics" a few months ago. As to the cost of plutonium, this is based very much on supply and demand. I think that the Oak Ridge people have done the most work on this and perhaps they can tell us some more about the philosophy of the cost of plutonium.

Dr. CAPRIOGLIO : I see at least one Oak Ridge man shaking his head to say no.

Dr. STEWART *(General Atomic)* : Well, let me just introduce Mr. Jaye on this subject. He did some of these studies once upon a time when he was at Oak Ridge, perhaps he can tell us something about it.

Mr. JAYE *(General Atomic)* : When I refer back to what I did while I was at Oak Ridge, it is rather a long time ago. The work has been going forward for the last six or seven years, on the value of all the fissionable materials both at Oak Ridge and at Hanford. Particularly plutonium has been studied extensively by Gene Eschbach at Hanford. He evaluated plutonium in much the same way as one would anticipate, namely, what is it worth relative to U^{235}. Many people concede that this is the proper way to evaluate the plutonium. The comment I would like most to make is relevant to Mr. Forbes-Gower's comment, about if we take plutonium away from the fast reactor, then we should pay what it is worth to the fast reactor and not what it is worth to a thermal reactor. I think that is the way I construe the question at least. I think that there are two comments on that : first, if one is going to pay what it is worth to a fast reactor, he has to look at when it is the fast reactor wants it. Namely, if the fast reactor is not ready for it until 1990 or perhaps 2000 or some number like that, then it turns out that if you apply negative working capital charges to this plutonium, it is worth almost nothing today. So on that basis, it is not the fast reactor price that determines the value of plutonium today, but rather the thermal reactor price. Now in the very, very long run, I think we come to the other end of the scale which Dr. Caprioglio was talking about, namely, perhaps in the year 2040 or 2050, when we install all of these nice fast reactors and they are producing plutonium at a huge rate, faster than the economy is growing, then they are looking for a customer. I think again it is going to be a question of supply and demand and there will be then a balance of thermal reactors or some other kind of reactor and high gain breeders.

I think that under these circumstances, we will find that it will be the user who will have a very large voice in what he will pay for his plutonium. And so it seems to me that at the two ends of the scale, the near term and the long term, we find the reactors users are determining the price of the plutonium and perhaps only during the intermediate range, whose length I would not like to guess, the fast reactor will indeed determine the price of plutonium. So, in summary, on the basis of what we have done, in the papers presented this afternoon, I think we properly evaluated the price of plutonium at the thermal reactor rate rather than the fast reactor rate.

Dr. CAPRIOGLIO : I think I would like to stress what you have said and make a comment.

Plutonium is going to be produced and sold and bought by those who are prepared to pay the highest price for it.

So that is why I think the high temperature reactor is interesting for plutonium burning, because we have got a feeling today that the high temperature reactor can pay the highest price for it. I do not see any other point in evaluating what the value of plutonium is, apart from the cost of recovering it from the spent fuel.

Mr. FORBES-GOWER *(CEGB)* : I am sorry to take the matter up again and I am not really wanting to conflict with you in taking it up again. The point was made earlier that there is a certain growth rate of electrical demand. The point was also made that there is some need to conserve fuel, though there are difference of opinion on how urgent this need is. The net result is that any public supply utility like the Board, cannot consider using plutonium other than in a symbiotic mode. I think that any consideration of parting with the plutonium, when we need it ourselves to sustain these doubling rates, is artificial. If the plutonium is being used in the symbiotic mode and the plutonium reactors are only to be considered as belonging to the same utility as produces the plutonium (— and personnally I think, this is just as true in other countries, I don't think, that the CEGB is alone in this —), I think that nearly all people who are going to use plutonium reactors will produce their own plutonium. In which case the costing arrangements will be very different and this goes back to the question of uncertainties. It is very apparent that the plutonium cost is not known to within 100 % or 200 %.

Mr. DOUGLAS *(Oak Ridge)* : I am sure that I will not help clarifying the confusion that exists on the price of plutonium. However, I thought I would mention a study that we performed at Oak Ridge relative to the Desalination Program. In this case, we were making an economic analysis of a very large reactor system and we were concerned about the monetary credit that one might take in anticipation of the sale of plutonium produced in a slightly enriched heavy water moderated reactor concept. In considering various aspects of the problem the question arose : If there is no market and nobody will buy the plutonium we are producing, what value is it to ourselves? So, although at the present time it is difficult to decide what is going to be the highest price paid for plutonium, we can at least set a minimum value. We arrived at this value by taking into consideration the additional penalty that one pays for processing and refabrication if one uses plutonium as an enrichment material and concluded that at a very minimum it is worth $3.50 per gram.

Mr. HOSEGOOD *(Dragon Project)* : It seems from what has been said that we are not too sure of the true future commercial cost of uranium-235. We have no idea whatsoever of the future cost or value of plutonium, so I think we should congratulate ourselves on how fortunate we are that everybody at present seems quite content to assume values which are very favourable to our type of reactor system in comparison with its competitors.

Dr. STEWART *(General Atomic)* : I think that is an overly pessimistic or cynical way to look at it. We have looked a little bit at how changes in separative cost would

affect the fuel cycle cost for various kinds of reactors, other of course than natural uranium reactors. We find that changes do not favour or penalize the HTGR system any more or less significantly than other systems. So I do not think that we need worry about favouring ourselves in this respect. I think that we worry too much also, if we feel that the prices of separated uranium are artificial. Certainly everything that I have been able to read on this subject seems to indicate that, if anything, the separative price of U^{235} will go down, not up.

Dr. CAPRIOGLIO : Any further comment or are we getting a bit tired all of us? Well, if this is the case, I would like to thank first of all, all the authors which have presented their papers to this Symposium and have allowed such a large amount of information to be distributed. I would also like especially to thank the rapporteurs, which have made quite a useful job, in trying to squeeze the juice out of all this amount of paper and pointing out to me the points that had to be discussed. Finally, I would like to thank all those who have participated in the discussion which has been as lively as a discussion can be. I would not like to terminate this Symposium without thanking Mr. Tytgat and all the secretariat which have done, I think, a good job out of it. So many thanks to everybody.

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